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DRYING AND STORAGE STUDY ON *ECHINACEA ANGUSTIFOLIA* ROOTS

BY

SHIYU LI ©

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The undersigned certify that they have read, and recommended to the Faculty of Graduate Studies and Research for acceptance, a thesis entitled **DRYING AND STORAGE STUDY ON *ECHINACEA ANGUSTIFOLIA* ROOTS** submitted by **SHIYU LI** in partial fulfillment of the requirements for the degree of **MASTER OF SCIENCE** in **FOOD SCIENCE AND TECHNOLOGY.**

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ABSTRACT

The suitability of forced air thin-layer dryer and fluidized-bed dryer for the dehydration of *E. angustifolia* roots was investigated by examining the drying characteristics of the roots, and evaluating the quality of dehydrated roots with echinacoside as a reference compound. Suitable storage conditions for preserving the echinacoside in ground dehydrated roots were also determined over a six- month storage study.

Two drying temperatures (50° and 30°C), air velocities (0.7 and 1.1 m/s for forced-air thin layer dryer, 3.3 and 4.8 m/s for fluidized bed dryer) and root sizes (small and medium) were examined in the drying experiments. Under similar drying conditions, the fluidized-bed dryer exhibited a higher drying rate than the thin-layer dryer at the initial drying stage. An extremely low drying rate was observed at the late stage of fluidized-bed drying, which might be attributed to the case hardening of the roots. Drying air temperature and root size were found to be important parameters affecting the drying rate in both thin-layer drying and fluidized-bed drying. Page equation appeared to fit the entire experimental data (R^2>0.96). Within the experimental range, equations correlating the experimental drying constants to air temperature, air velocity and size of roots were established. Thin-layer drying was better ($p<0.05$) able to preserve the echinacoside in the roots as compared to fluidized-bed drying. No significant ($p>0.05$) difference in echinacoside loss was observed between the two drying temperatures or air velocities specified in this study.

Echinacoside was satisfactory maintained in vacuum-packaged roots over 5 months of storage; however, a significant echinacoside decline was observed in the air-packaged roots after the 3rd month of storage. The effect of storage temperature on echinacoside change was minor ($p>0.05$). Therefore, room temperature storage might be acceptable for Echinacea products with respect to echinacoside preservation.

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1. INTRODUCTION AND LITERATURE REVIEW

1.1. GENERAL INTRODUCTION

Echinacea, a North American native herb, has long been traditionally used by the American Natives for medicinal purposes. It is currently adopted as a supportive therapy for cold, influenza, wound healing, chronic infections of respiratory tract or lower urinary tract, and chronic ulcerations (Bartels and Miller, 1998). The consumption of Echinacea products has been markedly increased both in Europe and North America during recent years. Numerous Echinacea products can now be found in natural health food stores or herb shops, in the forms of dry powder, capsules, tablets, liquid extracts, tinctures or injectable preparations.

Echinacea research is currently conducted in many countries including Germany, United States, Canada, Norway, Romania, Finland, Russia and New Zealand. According to Hobbs (1994), from the first laboratory work conducted in Germany in the 1930s to 1994, over 400 journal articles have been published on chemistry, pharmacology and clinical uses of Echinacea. The medicinal value of Echinacea species has been evaluated by recent scientific research. Laboratory and clinical studies on Echinacea in the past few years have demonstrated its immunostimulatory, antiviral and antibacterial benefits (Bauer and Wagner, 1991; Bodinet and Beuscher, 1991; Bodinet *et al.*, 1993; Parnharm, 1996). The scientific understanding on the active constituents on Echinacea species and their action modes

is still incomplete. However, it is known that among the many compounds that have already been identified, three categories of chemicals, i.e. lipophilic alkylamides, high molecular weight polysaccharides and caffeic acid derivatives are the most promising.

Public's increasing recognition of Echinacea has made it the most popular herbal remedy among both eclectic and regular medical doctors. For instance, Echinacea product was 131st on a list of the 2,000 most prescribed drugs in Germany in 1989 (Schumacher and Friedberg, 1991). According to a consecutive three-year survey conducted by Whole Foods Magazine, Echinacea was the best-selling medicinal herbal supplement in 1996 and 1997, accounting for the sale shares in natural food stores of 9.6% and 11.93%, respectively (Richman and Wikowski, 1997). The top 20 herbs being included in this survey are listed in Table 1.1. Natural products trading trends suggest that Echinacea and other medicinal herbal products will receive increasing attention. This is not only because of the scientific recognition of these products, but also because of consumers' conscious efforts at self-medication (Anonymous, 1997).

As a result of the large market demand, Echinacea has become the most widely and extensively harvested medicinal plant in recent years. In North America, most commercial Echinacea cultivation is in the western United States and Canada. The three most popular species are *Echinacea purpurea*, *E. pallida* and *E. angustifolia*. The *E. angustifolia* root, was the most frequently used part of Echinacea plants by indigenous Americans for both external and internal medicinal applications. Gilmore

(1913) stated that "applying the roots to areas of inflammation can relieve the sensation of burning by its feeling of coolness". The Lakota (Sioux) used the root and green fruit as a painkilling remedy for toothaches, tonsillitis, bellyache, pain in the bowel, or when one was thirsty or perspiring (Rogers, 1980; Munson, 1981). However, due to their biological nature, Echinacea roots deteriorate in a short time after harvest. To increase the usable economic life of freshly harvested roots, information on how to handle and preserve perishable roots after harvesting becomes very important. In addition, to make Echinacea root products available to consumers all year round, ensuring high quality roots during subsequent storage is essential as well. The present study attempted to provide some of this information.

Various technologies are available today for preserving biological products. Moisture control, primarily by drying, provides great potential for proper preservation of natural products. A traditional method of farm crop drying in most countries is sun drying, which is laborious, time consuming, and highly weather-dependent. With this method, a considerable loss, both in quality and quantity of crops is unavoidable. In accordance with the increasing applications of artificial farm dryers, many studies have been conducted to investigate forced heated air dehydration of agricultural products, with the objective of reducing the drying time and improving product quality (Giner and Calvelo, 1987; Jayas and Sokhansanj, 1989).

Heated air-drying is commonly applied to specialty crops, such as medicinal herbs or spices, because of its low cost and high capacity (Lebert *et al.*, 1992). The

need to safe-guard active constituents in medicinal herbs necessitates the use of lower temperature in the air drying process (Tabil *et al.*, 1999). However, the low temperature and usually relatively high moisture content in fresh herbal plants or their parts would contribute to a lower moisture removal rate. As a result, a drying process with long duration is usually expected. Therefore, selection of appropriate drying method and operating conditions (i.e. temperature, air velocity, air humidity) to effectively dry the herbs without damaging their medicinal properties demands more research attention.

Being a prized medicinal herb in North America, wild Echinacea has been harvested over 50,000 kg a year and shipped overseas for many years (Hobbs, 1994). However, information on how to propagate, cultivate, handle, and process the plant is notably lacking. Knowledge on drying and storage of Echinacea or parts thereof, and the resulting constituent changes in terms of medicinal quality is equally lacking. A better understanding of the drying and storage behavior of Echinacea roots in relation to their quality change under various conditions is necessary to develop an effective drying method and optimize the storage conditions. This project was thus initiated to rectify some of these deficiencies.

1.2. OBJECTIVES OF THIS THESIS

The primary objectives of the present research are as follows:

1. To investigate the suitability of forced air thin-layer dryer and fluidized-bed dryer in the dehydration of *E. angustifolia* roots. The study will:
   a. Determine the drying characteristics of the roots using the two air drying methods, and
   b. Evaluate the effects of the drying methods and drying parameters (temperature, air velocity, and size of roots) on the quality of the dehydrated *E. angustifolia* roots, with echinacoside as a reference quality marker.

2. To determine a suitable packaging and safe storage conditions for powdered roots. The study will monitor the change of echinacoside in vacuum or air-packaged powdered roots, stored at room or refrigerated temperature for six months.

1.3. LITERATURE REVIEW

1.3.1. Echinacea

1.3.1.1. Botany, Cultivation, and Medicinal Usage

Echinacea was originally used by the indigenous Americans to cure wounds, infections, and prevent insect and snake bites. It has numerous common names reflecting its use and appearance. Given in the order from the most to the least common, these names include purple coneflower, Echinacea, snakeroot, Kansas snakeroot, black sampson, narrow-leaved purple coneflower, scurvy root, Indian head,

comb flower, niggerhead, black susans, and hedgehog (Kindscher, 1989). There are nine species of Echinacea, namely, *E. laevigata, E. purpurea, E. paradoxa, E. atroubens, E. angustifolia, E. sanguinea, E. pallida, E. simulata, E. tennessensis*. All these species are indigenous to the United States (Chavez and Chavez, 1998). *E. angustifolia, E. purpurea* and *E. pallida* are the three most common species among the nine species of the genus Echinacea. Presently, only the roots and aerial parts of these three species are reported as being used for commercial supplies (Bauer and Foster, 1991).

Most of the world's supply of Echinacea is harvested from indigenous plants in the central United States. In Canada, it has been estimated that about 40 acres of Echinacea are planted in Alberta, 60 acres in Saskatchewan and 50 acres in British Columbia (Tabil *et al.*, 1999). There are two greenhouse growers of Echinacea with a total population of about one million seedlings in Alberta. Echinacea species are winter hardy and drought resistant (McGregor, 1968; Chapman and Auge, 1994). It can be propagated from seed, crown division, or by planting 10 to 12 cm root sections (Li, 1998). It produces stout, hairy, either single or branched stems 30 to 100 cm in height. Leaves are 15 to 30 cm long, ovate to lanceolate, rough and hairy. Depending on species, Echinacea root form varies from single taproot to fibrous (Awang and Kindack, 1991; Fostor, 1991). The root varies in size from the size of a pencil to a larger finger (Foster, 1991). Normally, the roots do not reach market size until 3 to 4 years after sowing, and they are usually harvested in the fall after the first frost has occurred (Li, 1998). Roots harvested in the fall have higher levels of active

ingredients and lower moisture than roots harvested in the spring (Blade, 1997). Information on harvest and post-harvest handling practices of Echinacea roots is very limited. As suggested by Blade (1997), plant parts above ground should be removed prior to root harvest and the harvesting can be done to a depth of 30-40 cm. As the newly harvested roots are usually able to last only for short periods at ambient temperature; cold storage after harvesting is necessary. According to Holm and Macgregor (1998), the recommended cold storage conditions for Ginseng, which are 1-5°C at 80 percent relative humidity (Van Dalfsen, 1992), should apply to most roots. Harvested roots should be washed and dried, using either forced air or placing them in an area of freely circulating air. The dried Echinacea roots are gray-brown or red-brown, wrinkled and twisted lengthwise, often in a spiral (Foster, 1991). The dried roots can be stored in a dry and cool area, and it is important to keep the roots out of direct sunlight. Blade (1997) also indicated that forced air-drying should not be performed at high temperature or over extended periods of time, but no temperature or time ranges were suggested.

The early medicinal usage of Echinacea has been extensively documented. According to Gilmore (1917), purple coneflower was "used as an antidote for snake bite and other venomous bites and stings and poisonous conditions" by all the American natives of the upper Missouri River region. In addition, Gilmore (1917) wrote, "Echinacea was used medicinally against more illnesses than any other plants". As cited by Bauer and Wagner (1991), Echinacea also appeared in several standard

works on medicinal plants, such as *King's American Dispensatory* (Felter and Lloyd, 1905) and *Webster's Dynamical Therapeutics* (Webster, 1898).

The widespread pharmaceutical recognition of Echinacea did not happen until the early 20th century. A German physician, H.F.C. Meyer, learned of its healing virtues from the American natives and introduced this plant to a professional remedy (Hobbs, 1994). However, due to the fact that the mechanism of Echinacea could not be explained in analogy with direct acting drugs, such as antibiotics, numerous clinical and laboratory studies were conducted on this medicinal herb in an attempt to understand the scientific facts on this herbal medicine (Stoll *et al.*, 1950; Wagner and Proksch, 1985; Wagner *et al.* 1988; Bauer and Wagner, 1991; Bodinet and Beuscher, 1991; Parnharm, 1996). Our current knowledge on this medicinal herb has been largely enhanced by these studies. It is known that Echinacea acts primarily via stimulation of the unspecific immune system. It is now possible to demonstrate the immunostimulatory activity of Echinacea extracts and assign the activity to certain groups of constituents found in the plant.

1.3.1.2. Chemical Constituents in Echinacea

Extensive research on chemical profile and physiological activity of Echinacea have been carried out since traces of alkaloid in Echinacea was isolated by Lloyd in 1897 (Hobbs, 1994). So far, the major groups of compounds identified in various Echinacea species include lipophilic alkylamides (Bohlmann and Hoffmann, 1983; Bauer and Remiger, 1989; Jakic *et al.*, 1994; Perry *et al.*, 1997), polysaccharides

(Stimpel *et al.*, 1984; Tubaro *et al.*, 1987; Wagner *et al.*, 1988; Luettig *et al.*, 1989; Roesler *et al.*, 1991; Steinmüller *et al.*, 1993), caffeic acid derivatives (Cheminat *et al.*, 1988; Bauer and Foster, 1991; Glowniak *et al.*, 1996; Pietta *et al.*, 1998), flavonoids (Bauer *et al.*, 1988), essential oil (Sayre, 1898; Sayre, 1903; Heyl and Staley, 1914; Neugebauer, 1949; Schulte *et al.*, 1967), polyacetylenes (Schulte *et al.*, 1967; Bauer *et al.*, 1987; Bauer *et al.*, 1988), and trace amounts of alkaloids (Lloyd, 1897). Culter (1930) was also able to measure the content of mineral ions in roots of *E. angustifolia*. Even though it is not possible to correlate the biological effects of Echinacea preparations to a specific chemical constituent, it is generally agreed that the immunostimulatory activity of Echinacea extracts depends on the combined action of lipophilic alkylamides, high molecular weight polysaccharides, and caffeic acid derivatives. Therefore, all these three classes of compounds might be used to standardize commercial Echinacea preparations (He *et al.*, 1998).

Alkylamides are a distinct class of natural compounds, containing an aliphatic acid (mostly unsaturated) residue linked with different amine moieties. The occurrence of alkylamides in Echinacea species was first reported by Jacobson (1954), who isolated a trace polyunsaturated alkylamide from the roots of *E. angustifolia* and later from *E. pallida* as well. This compound, being able to increase salivation in humans and exert a local anesthetic effect on the tongue, was identified by Jacobson (1954) as dodeca-(2E, 6Z, 8E, 10E)-tetraenoic acid isobutylamide. Other new alkylamides were found thereafter in *E. angustifolia* and *E. purpurea* (Bohlmann and Grenz, 1966; Verelis, 1978). According to the recent studies by Bauer and Remiger (1989), eleven

alkylamides were found in *E. purpurea* roots, some of them were also present in the leaves (Bauer *et al.*, 1988). In the roots of *E. angustifolia*, total of 15 alkylamides were identified, with main constituents being isomeric dodeca-(2E, 4E, 8Z, 10E/Z)-tetraenoic acid isobutylamides (Bauer *et al.*, 1988). Only a very small quantity of alkylamides was found in *E. pallida* roots compared to those contained in *E. angustifolia* and in *E. purpurea*.

Choné (1965) described the occurrence of a polysaccharide fraction in Echinacin®, a freshly expressed juice from the aerial parts of Echinacea plant, but presented no data on its structure. A polysaccharide mixture with weak antihyaluronidase activity, consisting mainly of an acidic mucopolysaccharide was isolated by Bonadeo *et al.* (1971). Two immunostimulatory polysaccharides (PSI and PSII) were isolated from the aerial parts of *E. purpurea* (Proksch, 1980; Wagner and Proksch, 1981). Studies showed PSI to be a 4-O-methyl glucurono-arabinoxylan, while PSII was shown to be an acidic arabinorhamnogalactan. Polysaccharides have recently been obtained from cell cultures of *E. purpurea* as well (Wagner *et al.*, 1988). The high molecular weight polysaccharides in Echinacea have recently been demonstrated to activate a broad variety of cytotoxic and secretory functions of murine and human macrophages *in vitro* (Stimpel *et al.*, 1984; Luettig *et al.*, 1989; Roesler *et al.*, 1991).

The caffeic acid derivatives in Echinacea species involve echinacoside, cynarin, cichoric acid, chlorogenic acid, etc. These compounds have recently been

proven to have a strong protective effect on the free-radical-induced degradation of collagen, which mimics free radical-mediated skin damage by UV (Facino *et al.*, 1995). The protective potency by these compounds was shown to be in the following order: echinacoside ≈ chicoric acid > cynarin ≈ caffeic acid > chlorogenic acid.

The quinic acid derivative cynarin (1, 5-O-dicaffeoylquinic acid), identified by Bauer and Wagner (1987) as a characteristic constituent of *E. angustifolia* roots, is potentially useful for differentiating *E. angustifolia* from *E. pallida*. Cichoric acid (2-3-O-dicaffeoyltartaric acid) was first isolated from *E. purpurea* (Hsieh, 1984). It is especially abundant in the flowers and roots (1.2-3.1% and 0.6%-2.1%, respectively), and much less is present in the leaves and stems (Bauer *et al.*, 1988). The optical activity of cichoric acid isolated from *E. purpurea* was described by Remiger (1989) and Soicke *et al.* (1988). The presence of chlorogenic acid (3-O-caffeoylquinic acid) and isochlorogenic acids in the leaves and stems of *E. pallida* and *E. angustifolia* was first demonstrated by Bauer *et al.* (1988). Cheminat *et al.* (1988) identified the isochlorogenic acids of *E. pallida* as 3,5- and 4,5-O dicaffeoylquinic acids.

Echinacoside (also called phenolic acid and/or tri-saccharide) was first isolated from the roots of *E. angustifolia* by Stoll *et al.* (1950). They demonstrated that the activity of 6.3 mg of echinacoside in 8×10^{-3} molar solution against *Staphylococcus aureus* was equivalent to about 10 Oxford units of penicillin. Eilmes (1976) provided evidence for interferon-activity in relation to the echinacoside isolated from *E. angustifolia*. The structure of echinacoside is complex and unusual, as can be seen

from Figure 1.1. It contains two molecules of glucose and one molecule each of rhamnose, caffeic acid and catecholethanol. The linkage of the five components was determined by Becker (1982). In the meantime, echinacoside has also been found in *E. pallida* (Bauer and Wagner 1987), *E. simulata, E. paradoxa,* and *E. atrorubens* (Bauer and Foster, 1991).

Echinacoside is commonly used as a reference standard for some European Echinacea products (Hobbs, 1994). Although some studies (Schumacher and Friedberg, 1991; Foster, 1991, Bauer, 1998) expected that this compound is probably not particularly active, echinacoside is still recognized as a good candidate for Echinacea quality standardization as it is unique to Echinacea species and accumulates especially in the roots. Compared to alkylamides or polysaccharides, echinacoside can be easily identified with the aids of high performance liquid chromatography (HPLC) and thin layer chromatography (TLC) techniques. Therefore, it was selected as the quality marker for *E. angustifolia* roots in the present study. According to recent investigations, *E. pallida* roots contain 0.4-1.7% (on a dry weight basis) and *E. angustifolia* roots 0.3-1.3% echinacoside (Bauer and Remiger, 1989). Echinacoside was also recently found in the flowers of *E. angustifolia* (0.1-1%) and *E. pallida* (traces) (Bauer *et al.*, 1988). *E. purpurea* contains undetectable level of echinacoside (Bauer and Wagner, 1991; Hu and Kitts, 2000). The aerial parts of *E. angustifolia* and *E. pallida* were recently shown to contain verbascoside, a structural analogue of echinacoside, containing only one molecule of glucose (Bauer *et al.*, 1988).

1.3.2. Drying Process

1.3.2.1. General Principles of Drying

Drying involves the removal of a liquid, usually water, from a solid by thermal means. The main technical or commercial objectives of drying are to extend storage life, enhance quality attributes, such as color, palatability or digestibility, ease of handling, or to assist further processing.

Two important physical phenomena, i.e. heat transfer and mass transfer, occur simultaneously throughout a drying cycle. Heat transfer raises the temperature of wet solid and evaporates the moisture. The heat is transferred from a heating medium to material through convection, conduction, radiation or any combination of these mechanisms. Mass transfer, primarily the movement of moisture, is a function of pressure differential between the vapor in the product and the vapor in the surrounding atmosphere (Andales, 1981). When a product containing bound moisture is placed in heated air of specific temperature and humidity, it will lose or gain moisture until the vapor pressure of the moisture within the product becomes equal to its partial pressure in the air. This moisture content, which is dependent on the humidity and temperature of the air, is termed equilibrium moisture content.

A drying process is often analyzed based on the drying curves, which could be obtained by plotting the data (e. g. moisture ratio and drying time) collected from the drying trials. In general, drying of different materials under different but constant conditions could often give curves of different shapes. However, three major drying

stages, namely, constant rate drying stage, first falling rate drying stage, and second falling rate drying stage, can usually be observed on a drying curve in a convection drying process (Figure 1.2) (Threlkeld, 1965). In the constant rate period, drying takes place from the surface of the material and is similar to evaporation of moisture from a free water surface. This period continues as long as water is supplied to the surface as rapidly as evaporation takes place. The constant rate period ends when the critical moisture content is reached, and the drying enters the first falling rate period characterized by a decreasing drying rate due to a decreasing area of wet surface. As drying proceeds, the fraction of wet surface decreases to zero, at which point the second falling rate drying begins and continues until the equilibrium moisture content is reached.

The dryers used in industry can differ in various applications and designs, but in general, they will fall into four main categories according to the principal heat transfer method employed. They are convection dryers (direct dryers), e.g. spray dryer, thin-layer dryer, and fluidized-bed dryer; conduction dryers (indirect dryers), e.g. drum dryer, vacuum dryer; dieletic dryers and some specific dryers such as freeze dryer.

1.3.2.2. Drying of Agricultural Crops

Drying has become increasingly an important part of agricultural production systems (Hall, 1980). Agricultural products drying is carried out primarily to ensure stability of product quality for a given storage period or to ensure product availability

on a year-round basis. Traditional open-air sun drying is still the most common drying method for crops in the field (Yaciuk, 1981). With "cost-effectiveness" as an advantage, there are obvious disadvantages associated with sun drying, which often results in a poor quality of the dried product: no control over the drying process; possible contamination of the product by dirt, rodents, and animals; infestation by insects or molds (dried too slow); and exposure of the product to rain and wind, which causes repeated rewetting and redrying. Artificial heated air-drying permits a quick removal of the moisture in crops. However, due to a greater expense involved with a heated air system, and the possibilities of fire hazard and crop damage if a high temperature is used, this technique became widely practiced on farms only decades ago (Hall, 1980). Over recent years, there have been attempts to concentrate on the development of forced heated air dryers that can be effectively used in agricultural activities.

Air drying of farm crops, which are biological in nature, differs from drying of other organic or inorganic materials, such as paper, sand, chemicals, etc. Firstly, the constant rate drying period is short in duration for farm crops. Therefore, the falling rate interval is the most important period from the standpoint of drying of harvested crops. Even when the constant rate period is in effect at the start of drying, this period is often neglected by researchers because of its short duration and the small amount of moisture being removed before entering the falling rate period. Secondly, although the external conditions and the internal mechanisms are both important in controlling farm crop drying, the internal mechanism of moisture movement is more fundamental. This

indicates that the biological characteristic of crop itself could be a very influential factor on the drying process. Therefore, to better understand the drying behavior of a specific agricultural crop and to design efficient dehydrators, appropriate support models for prediction and optimization of process parameters are required.

Based on the review of Tabil *et al.* (1999), among the numerous drying studies on plants or crops, the following five models were popularly used for fitting the data obtained from the drying of biological materials:

1. Exponential model (Lewis, 1921)

$$MR = e^{(-Kt)} \tag{1}$$

2. Page equation (1949)

$$MR = e^{(-Kt^{n})} \tag{2}$$

3. Thompson *et al.* equation (1968)

$$t = A \ln(MR) + (B \ln(MR))^2 \tag{3}$$

4. Approximation of diffusion model

$$MR = Be^{-At} \tag{4}$$

5. Two term exponential equation

$$MR = A_0 e^{-K_1 t} + A_1 e^{-K_2 t} \tag{5}$$

Where:

MR = moisture ratio, determined by the following equation:

$$MR = \frac{M - M_e}{M_0 - M_e}$$

A_0, A_1, A, B, K, K_1, K_2, n = experimentally determined constants

t = time (min)

M_0 = initial moisture content (% d. b.)

M = moisture content (% d. b.)

M_e = equilibrium moisture content (% d. b.)

Among these models, the equation developed by Page (1949) has been extensively used by many researchers to characterize the thin-layer drying of agricultural products. The crops investigated in those studies included soybean (White *et al.*, 1981), barley (Jayas and Sokhansanj, 1989), American Ginseng (Li and Morey, 1987), kenaf (Muhidong *et al.*, 1992), rapeseed (Pathak *et al.*, 1991), alfalfa (Patil *et al.*, 1992), etc. Even though higher air velocities are usually applied in fluidized-bed drying and the dried products are usually small particles, the equation by Page (1949) was found to be applicable to this drying procedure (Prasad *et al.*, 1994; Swasdisevi *et al.*, 1999).

1.3.2.3. Effects of Drying on Chemical Constituents in Specialty Crops

The processed product is rarely nutritionally superior to the raw ingredients from which it is produced as nutrients are usually destroyed during processing because

of sensitivity to pH, oxygen, light, heat, or a combination of these factors (Kramer, 1982). Different drying processes may have different impacts on the chemical constituents of the plants. For example, more than five times as much carotene content in alfalfa can be preserved with artificial drying as compared to sun drying (Dimattia, *et al.*, 1996). Kaminski *et al.* (1986) reported that the loss in volatiles of carrots was 69% by freeze-drying, 75% by hot air-drying, 81% by fluidized-bed drying, and 84% by microwave hot air-drying, respectively.

In general, better preservation of nutrients will be found when lower dehydration temperatures and shorter drying times are employed. Freeze-drying technology, which is based on sublimation of frozen moisture from the material under vacuum, allows the lowest possible temperatures used in dehydration. Moreover, the vacuum applied in this technique would minimize oxidative processes, leaving the product relatively unaffected with respect to its physical and chemical structures (King *et al.*, 1989). Therefore, freeze-drying has often been used as a method for drying samples to be used for further chemical analysis (Tsourouflis, 1976). Also, freeze-drying is regarded as the most gentle method to preserve plant materials for analysis of various organic substances (Popp *et al.*, 1996).

The major drawbacks of freeze-drying are its energy cost and the lengthy drying period, which prohibit its large-scale application (Liapis *et al.*, 1996). Tabil *et al.* (1999) reviewed the drying methods used in dehydration of specialty crops and found that heated forced-air drying was most commonly employed. However, herbs or

spices can lose some of their constituents during air dehydration. Oxidation is the primary cause of such a loss in this process, but non-oxidative loss, such as heat or enzymatic degradation could occur as well.

Mechanisms for constituent loss in heated-air drying can be quite variable. In general, important mechanisms primarily depend on a number of physical factors that affect the chemical degradation rate of constituents. Very often, the physical factors influencing the drying rate are those affecting the rate of constituent loss. These include drying temperature, moisture content of the product, air velocity, and relative humidity of the air. Therefore, to ensure the maximum retention of useful constituents in herbal products during heated-air drying, proper selection of operation parameters is critical. Extensive studies have been conducted to evaluate the behavior of volatile aromatic compounds and related quality changes in herbs or spices during drying (Huopalahti *et al.*, 1985; Pääkkönen *et al.*, 1990a, 1990b; Lebert *et al.*, 1992; Raghavan *et al.*, 1994; Rocha *et al.*, 1993; Venskutonis *et al.*, 1996). It was found that the extent of changes in the amount of desired compounds depend on the drying method, temperature, velocity and humidity of air as well as the biological characteristics of the herbs and spices.

In the dehydration of medicinal herbs, the operational conditions are even more crucial because improper drying could damage biologically-active constituents in the plant, leading to the loss of most or all of the medicinal value. American Ginseng is an economically important medicinal plant mainly grown in the northern

U.S. According to Reynolds (1998), optimum drying temperature for preserving active constituents of American Ginseng was from 32 to 38°C. Drying temperatures below 30°C can result in mold growth during drying, but temperatures above 40°C may lead to an undesirable brown internal color and severe loss of active constituents. The steroidal alkaloid, solasodine from the plant *Solanum laciniatum,* is a potential raw material for steroid drug manufacture. Solasodine occurs in the plant as the triglycosides, solasonine and solamargine. It was reported by Crabble and Fryer (1982) that the *Solanum* leaves could be well dried either at ambient conditions or below 100°C (<50°C was recommended). Drying above 100°C causes severe loss of solasodine unless it is used only for the early stages of water removal.

Not much work has been reported on the effects of drying on Echinacea components. According to Bauer and Wagner (1991), the polyacetylenes and cichoric acid are quite unstable and might not be retained in commercial Echinacea products. Recently, Perry *et al.* (1999) focused their work on the changes in alkylamides during processing, drying and storing of *E. purpurea* roots. Their data indicated that chopping process could alter the levels of some alkylamides slightly, whereas 32-33°C oven drying for 48 h had no effect on alkylamide levels in the roots.

Different compounds could behave quite differently in the drying process. The influence of heated-air drying on the composition of both volatile and non-volatile flavour compounds of different Dutch bell pepper cultivars was investigated by Luning *et al.* (1995). It was found that the contents of glucose, fructose, ascorbic acid,

citric acid, and oxalic acid were significantly decreased by drying, but the levels of sucrose, malic acid, fumaric acid and *cis*-aconitic acid were increased. To date, no data on the stability of echinacoside during drying and storage exists. However, with phenolic group in its structure, echinacoside should theoretically behave somewhat similar to other phenolic compounds when it is subjected to heat. According to De Moura and Dostal (1965), chlorogenic acid loss was observed during the drying process of various prune cultivars. Phenolic compounds in plums were also found to be modified during drying and in particular in the initial drying stages (Moutounet, 1978).

Our current knowledge on the stability of echinacoside is largely limited to the process of analytical method development for quantitation of echinacoside. When conducting an extraction recovery test, 1 h Soxtech extraction with 70 mL methanol yielded the best recovery of echinacoside, suggesting that echinacoside in a solution could resist heating at 64.6°C (boiling point of methanol) for a maximum of 1 h. A higher temperature or longer heating time could cause severe loss of echinacoside. Although the mechanism remains unclear, preliminary tests indicated that frozen roots would lose most echinacoside if air-drying were employed. It has been hypothesized that upon thawing, before and during air drying, enzymes responsible for hydrolysis of phenolic compounds would be released causing the loss of echinacoside. During drying, tissues damaged by freezing and thawing would also expose the compounds, causing further loss through oxidation.

Based on the above information, selection of drying method and related operating parameters can be critical to the preservation of the quality and the cost of drying of valuable plants.

1.3.3. Post-drying Storage of Specialty Crops

Much research on post-harvest storage of fresh plants has been conducted in an attempt to effectively extend their storage life (Weichmann, 1986; Bracy, *et al.*, 1992; Sankat *et al.*, 1995; Patil *et al.*, 1995; Jeon and Lee, 1999). It is very important as well to have storage data on dehydrated herbs or spices, since even though these products are essentially inert, the storage losses can occur through both chemical deterioration and microorganism or insect attack.

The microorganisms harbored by most dehydrated herbs are aerobic sporeformers and molds (Malmsten *et al.*, 1991). These organisms are often responsible for generating heat and moisture in stored bulk materials, hence reduce the value of herbs by adversely affecting their flavor, enhancing chemical reactions and producing toxic metabolites. Though the storage loss caused by molds or insects is the main problem for herbs, the value loss of processed herbal products due to chemical degradation taking place during storage can not be ignored. According to Baritaux *et al.* (1992), the concentration of most odor components in basil was reduced after 7 months of storage. Moreover, freeze-dried summer savory stored under oxygen atmosphere lost aroma compounds even during a short storage period (Pääkkönen *et al.*, 1990a). In terms of herbal products for medicinal purpose, the modifications of the

biologically-active chemicals should be avoided by all means during storage as it is the final step prior to marketing the product. The loss of active constituents during this period can, therefore, exert a major influence on the ultimate quality of the products. Unfortunately, scientific information on optimal storage of processed medicinal plants is very scarce.

Chemical changes during storage are largely dependent on the nature of the product and the storage conditions such as length of storage, temperature, and atmospheric oxygen. In general, the nutritional value of a stored material is degraded when it is subjected to either oxidation, light, heat, or pH shift (Heidelbaugh and Karel, 1982). Heat is undoubtedly the prime cause of chemical losses, particularly ascorbic acid, thiamin, folic acid, and lysine (Kramer, 1982). Therefore, the best preservation from chemical degradation is to use low storage temperature. According to the storage study with basil, marjoram and wild marjoram (Pääkkönen *et al.*, 1990b), the flavor and odor of those dried spices were adversely affected when the storage temperature was elevated from 23 to 35°C.

Packaging is most frequently used to protect the stored products from changes due to moisture absorption, oxidation, and light. Materials used in food and drug packaging include glass, metals, paper, wood, and plastics such as cellophane, polyolefins, vinyl derivatives, polyesters, rubber hydrochloride, and ionomers (Heidelbaugh and Karel, 1982). Low-cost polyethylene film can be used for packaging of dried herbal products. However, it has the disadvantage of being very permeable to

oxygen and water vapor so that it provides very little protection to the package contents. Most dehydrated herbal products, particularly when they are in powder form, are highly hygroscopic. Therefore, airtight containers or multi-films which offer the advantages of attractiveness, toughness, and good barrier characteristics are always preferred for prolonged storage with respect to nutrient preservation. Some specific herb products being sensitive to oxygen might be packed in nitrogen atmosphere or vacuum. According to Pääkkönen *et al.* (1990b), the intensity of odor and taste of freeze-dried basil was better preserved in vacuum and nitrogen-filled packages than in paper bags at room temperature. Moreover, medicinal materials requiring protection from light should be kept in a light-resistant container. Alternatively, the container may be placed inside a suitable light-resistant covering and/or stored in a dark place.

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Table 1.1. Herbal supplement sales in health food stores[1]

'97 rank	Herb	'97 % of sales	'96 % of sales	'96 rank
1	Echinacea	11.93%	9.6%	1
2	Garlic	8.52%	7.2%	2
3	Ginkgo biloba	6.80%	5.1%	4
4	Goldenseal	5.95%	4.7%	5
5	Saw palmetto	4.87%	3.1%	9
6 (tie)	Aloe	4.76%	2.4%	12
7 (tie)	Ginseng	4.76%	6.4%	3
8	Cat's claw	3.49%	2.1%	14
9	Astragalus	3.07%	1.3%	27
10	Cayenne	2.83%	2.5%	11
11	Siberian ginseng	2.70%	3.5%	7
12	Bilberry	2.61%	1.6%	23
13	Cranberry	2.47%	1.7%	18
14	Dong quai	2.13%	1.8%	17
15	Grape seed extract	2.07%	2.0%	15
16	Cascara sagrada	1.92%	2.8%	10
17	St. John's wort	1.87%	N/A	N/A
18	Calerian	1.73%	2.2%	13
19	Ginger	1.69%	1.7%	18
20	Feverfew	1.59%	1.6%	23

* Four herbs (cat's claw, saw palmetto, ginkgo biloba, and echinacea) have been on the list all three years that the study has been conducted. The top 10 herbs have 56.98% of sales; top five herbs have 38.07%, second five herbs have 18.91%.

[1]Adapted from Richman and Witkowski (1997).

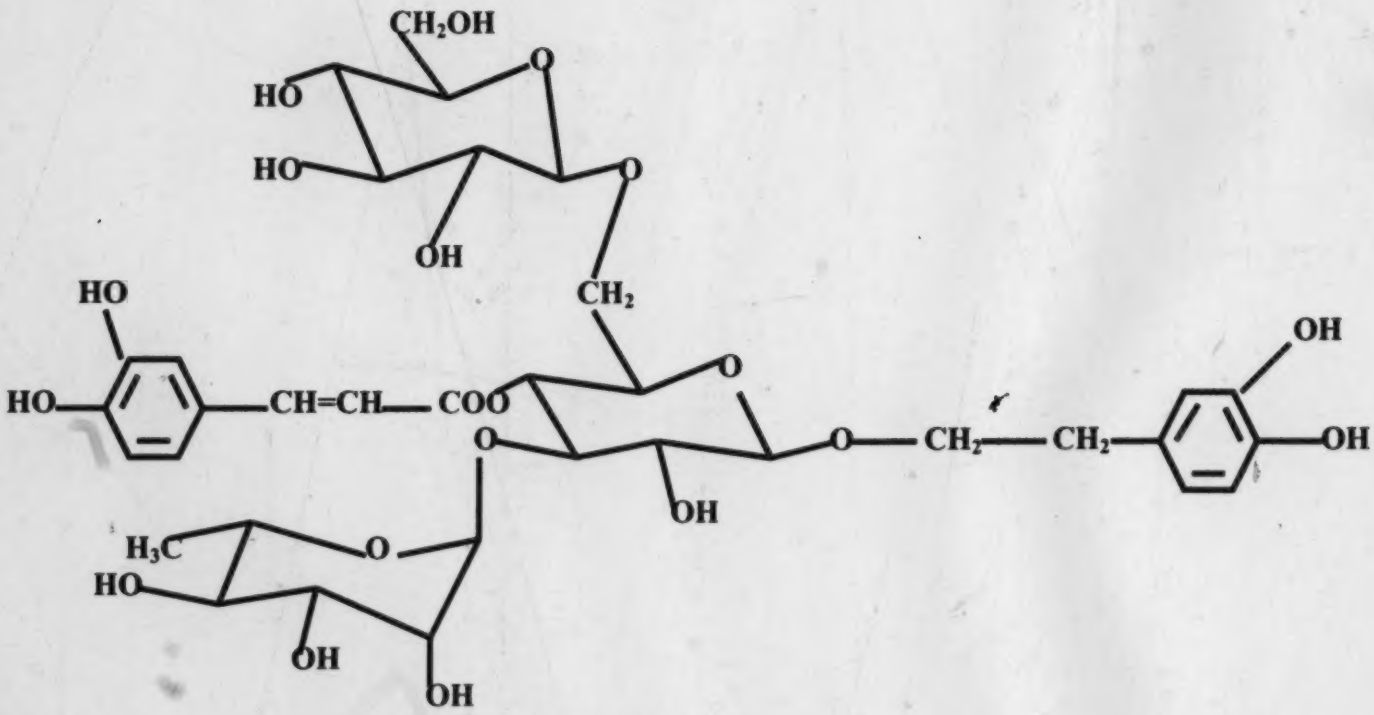
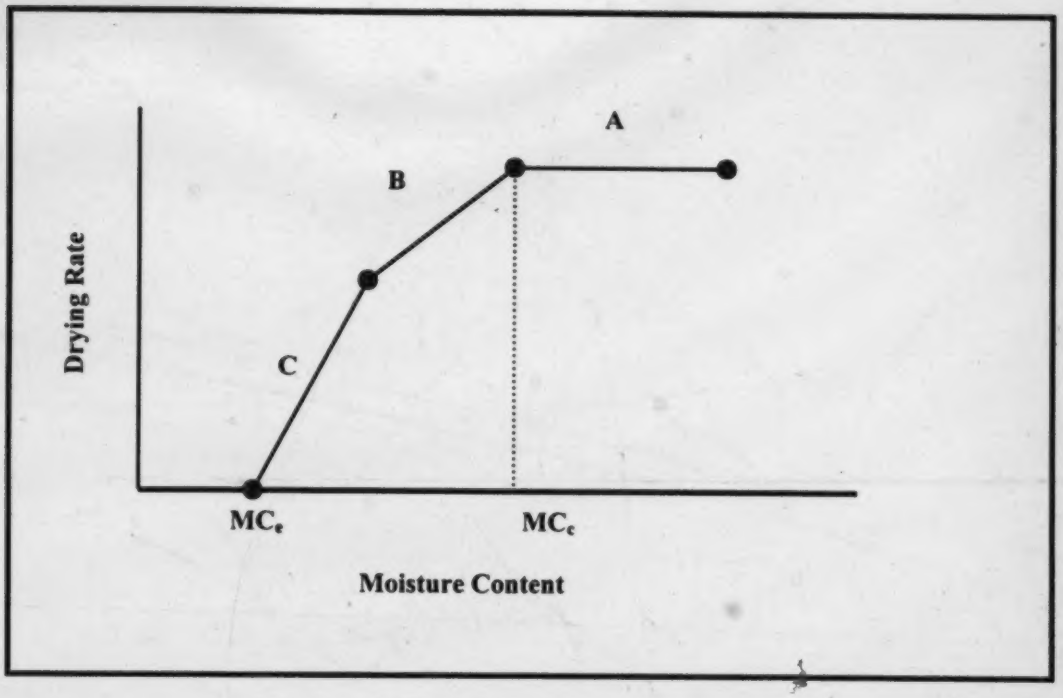


Figure 1.1. Chemical structure of echinacoside (Stoll *et al.*, 1950)



A: Constant rate drying B: First-falling rate drying C: Second-falling rate drying

MC_c : Critical moisture content MC_e : Equilibrium moisture content

Figure 1.2. Drying rate for a wet product under constant drying conditions (Threlkeld, 1965)

2. DRYING STUDY ON *E. ANGUSTIFOLIA* ROOTS

2.1. INTRODUCTION

Although research information on drying of *E. angustifolia* roots is sparse, one can always develop an effective drying method with appropriate operation parameters following similar strategies used for other herbs and medicinal plants. According to the study on twenty-seven types of Chinese medicinal herbs by Chu *et al.* (1992), as cited by Tabil *et al.* (1999), variable temperature drying is the best for optimizing heat and mass transfer conditions. A proper air flow rate is necessary as well to transfer heat for water evaporation to the material and to carry away evaporated water. Experiments on ginseng drying conducted by Van Dalfsen *et al.* (1992) showed that at a given drying temperature, ginseng roots reached 35% of their original weight in about 7.5 days at an air speed of 0.2 m/s, while at 0.5 m/s it took only 5.5 days. The proper relative humidity of drying air was also proven to be very crucial to a drying process (Tschernitz and Simpson, 1979; Rocha *et al.*, 1993).

In evaluating the feasibility of a dehydration method and related operational conditions, both the economic factors, such as drying time, and the quality of final product must be considered. In the case of natural biological products, it is normal for quality considerations to override economic factors (Baker, 1997). For example, important quality measures of ginseng roots are contents of polysaccharides and

ginsenoside, a complex mixture of compounds called triterpene saponins. Reynolds (1998) found that the concentration of malonyl ginsenosides was decreased if drying temperature was increased from 38 to 44°C. Total ginsenoside concentration can be reduced by 26% at a drying temperature of 44°C and by 17% at 38°C as compared to freeze-dried root. Similarly, 38°C was concluded by Reynolds (1998) as the optimum drying temperature for the preservation of both chemical and physical quality of American ginseng roots.

The immunostimulant activity of Echinacea may have been related to polysaccharides, polar caffeic acid derivatives, and/or lipophilic alkylamides, or most likely the combination of these three groups of compounds. Therefore, the quality standardization of Echinacea roots after processing becomes very essential. Perry *et al.* (1999) suggested that alkylamides could be one of the classes of compounds most relevant to standardization of Echinacea preparations. However, the unequivocal identification of alkylamide peaks requires isolation of the individual compounds, necessitating multiple purification steps as well as mass spectrometry (MS) and nuclear magnetic resonance (NMR) analysis, resulting in increased analytical complexity and cost. Caffeic acid derivative echinacoside, the unique component for most Echinacea species, is commonly used as a quality reference marker for Echinacea products (Hobbs, 1994). In Alberta, the concentration of echinacoside is normally used to determine the price of *E. angustifolia* roots (Hoyano, 1999). The Agri-Food Laboratory of Alberta Agriculture, Food and Rural Development (AAFRD), has been working on isolation and identification of echinacoside. They have developed a rapid method for extraction,

isolation, purification, identification, and quantitation of echinacoside. For the present study, echinacoside was selected as the reference quality marker throughout the drying and storage experiments.

In this study, the suitability of two methods, i.e. thin-layer air drying and fluidized-bed drying was evaluated. Thin-layer air drying refers to the drying process of a product, which is entirely exposed as a single layer to the air moving over the product. Depending upon the air conditions in the vicinity of the thin layer, the amount of moisture removed from the product can be determined by a thin-layer drying equation applicable in the range of air conditions. The thin-layer model could be used to accurately predict drying procedures in deep beds. A considerable amount of work has been reported in the literature on thin-layer drying of crops (Hukill and Schmidt, 1960; Ross and White, 1972; Wang and Singh, 1978; Misra and Brooker, 1980; Syarief *et al.*, 1984; Noomhorm and Vera, 1986; Muhidong *et al.*, 1992). The roots of American ginseng harvested on three different dates were comparatively dried in a thin-layer dryer at three temperatures (27, 38, and 49°C), two air flow rates (0.5 m^3/ s m^2 and 0.3 m^3/ s m^2), and two sizes of roots. The drying rate under each specific drying condition was evaluated by Li and Morey (1987). It was noted that drying air temperature had the greatest impact on drying rate, particularly when moisture contents were below 28 to 30% (w.b.). The diameter of the roots had some effect whereas air flow rate and initial moisture content had no obvious effect on drying rates. In addition, it was found that the cold storage prior to drying of ginseng roots did not decrease the root quality in the subsequent drying (Van Dalfsen *et al.*, 1992).

Fluidized-bed drying technique is another one of the most important modern drying methods. It uses hot air to dry a material in which the bed of the material is expanded by applying a critical air velocity and reaches a state similar to a boiling liquid. The literature is lacking on the use of fluidized-bed technique to dehydration of medicinal plants. The high thermal efficiency and uniform temperature throughout the bed of material could make fluidized-bed drying an attractive choice over several conventional techniques for herb drying. It has been reported that in comparison with stationary-bed drying, fluidized-bed drying provided 15% and 54% higher drying rate on grain at a low (20°C) and a high (65°C) temperature, respectively (Uckan and Ulku, 1986). According to Treybal (1980), large air volumes must be blown at a high velocity of 3 to 4 m/s to overcome the non-uniformity of moisture distribution in the product taken from various parts of the dried sample. More scientific data are required to support the application of this drying method to medicinal plants.

Therefore, the objectives of the current study are:

a) to determine the drying characteristics of *E. angustifolia* roots in forced air thin-layer dryer and fluidized-bed dryer by analyzing the drying profiles of the roots, and;

b) to evaluate the effect of the drying methods and drying parameters (temperature, air velocity, and root size) on quality of dehydrated *E. angustifolia* roots, with echinacoside as a reference quality marker.

2.2. EXPERIMENTAL DESIGN

To achieve the objectives of the study, drying experiment was divided into two parts:

Part A. Drying *E. angustifolia* roots in forced-air thin-layer dryer and fluidized-bed dryer under various drying parameters. Under each set of drying conditions, two replicate runs were performed. The drying profiles of *E. angustifolia* roots under various conditions, with respect to moisture ratio, rate of moisture removal, etc., were determined. Drying models for *E. angustifolia* roots in both thin-layer and fluidized-bed dryers were developed. The experimental variables used were:

Thin-layer drying:

a) Drying temperatures: 30° and 50°C
b) Air velocities: 0.7 and 1.1 m/s
c) Root sizes: small (diameter<3 mm) and medium (diameter 3-5.99 mm)

Fluidized-bed drying:

a) Drying temperatures: 30° and 50°C

b) Air velocities: 3.3 and 4.8 m/s

c) Root sizes: small (diameter<3 mm) and medium (diameter 3-5.99 mm)

Part B. Dried root samples obtained at each of the above experimental conditions were analyzed by reverse phase high performance liquid chromatography (RP-HPLC) with echinacoside as the chromatographic marker. HPLC analysis of the sample from each drying treatment was performed in duplicate. The root quality under each drying condition was accordingly evaluated, and a more suitable drying method and drying conditions were selected and recommended.

2.3. PART A: DRYING CHARACTERISTICS STUDY ON *E. ANGUSTIFOLIA* ROOTS

2.3.1. MATERIALS AND METHODS

2.3.1.1. Treatment of Raw Roots

Fifteen kilograms of field-grown *E. angustifolia* roots from 3-year old plants were transported from Crop Diversification Centre-South (CDC-South) to University of Alberta in coolers within 6 h. As reported by Van Hooren and Lester (1989), the ginseng

roots being refrigerated prior to washing and drying were more desirable than roots washed and dried directly after harvest. According to Reynolds (1998), the optimum temperature for post-harvest ginseng roots conditioning was between 3°C and 8°C. Within this temperature range, minimum dry matter loss induced by respiration occurred. Based on such information, fresh *E. angustifolia* roots were stored at 4°C overnight upon their arrival. The root crowns and all decayed or damaged parts were removed completely. On the following day, the roots were washed in cold water to remove soil and rinsed with clean water. Clean roots were then spreaded on paper towels, and left for approximately 3-4 h at room temperature until surface moisture evaporated. The two-end diameters of each individual root were measured using an electronic caliper with a two decimal resolution. The mean diameter of individual root was determined by averaging the two-end diameters. Roots were then sorted into two categories as follows: a) mean diameter >3.00 mm as small roots, and b) mean diameter 3.00-5.99 mm as medium roots. The clean, undamaged, disease-free, and sorted roots were modified atmosphere packaged (MAP) in 3 mil low-density polyethylene (LDPE) bags OF 200 g-size. The bags were sealed by Multivac packaging machine (AG500, Sepp Haggenmüller KG, Germany). The gas mixture used for MAP containing 25% carbon dioxide, 3% oxygen and 72% nitrogen was provided by a proportional tri-gas blender (180 SCFH, Smith Equipment, Watertown, SD). These MAPed roots were finally transferred in a cooler to Agricultural Value-added Engineering Centre (AVEC) of Alberta Agriculture, Food and Rural Development, and stored at 4°C until further processing.

2.3.1.2. Drying Apparatus

The drying experiments were carried out using a laboratory forced-air thin-layer dryer and a fluidized-bed dryer. The schematic diagrams of those dryers are shown in Figures 2.1. and 2.2., respectively.

2.3.1.2.1. Prototype of fluidized-bed dryer

A lab-scale fluidized-bed dryer was developed and built at the University of Alberta. The fluidized-bed dryer consisted of a cylindrical acrylic drying chamber, an air velocity reducer, a constant pressure compressed air supply, an electrical heating unit, a thermometer, and a voltage transformer. The acrylic drying chamber, 51 mm in diameter, 3.2 mm wall thickness and 610 mm in height, was fitted with a 101-51 mm PVC air velocity reducer (plumbing fitting) on top. Constantly supplied compressed air (100 psi) was used as the drying medium. A control valve and a gas regulator, both manually operated, were attached at the outlet of compressed air so as to regulate the air flow through the system at a desired level. The compressed air was heated by an electric air heating unit before it entered the drying chamber. Voltage supply to the heater was regulated through a manually controlled transformer (3PN1010, Staco Inc., Dayton, OH), by which a desired air temperature was achieved. A thermometer with 1°C accuracy was used to monitor the inlet air temperature. Heated air entered the drying chamber by passing through a 13 mm hose, which was connected to the chamber with a quick-coupling junction. A group of drinking straws placed between the air inlet and the 51 mm PVC fitting were used to straighten air stream before it reached the product. Air velocity

in the chamber was measured at a port 76 mm below the reducer by a hand-held hot-wire anemometer (P-37000-52, Labcor, Québec).

2.3.1.2.2. Forced air thin-layer drying facility

The forced air thin-layer dryer was designed and constructed by the engineers at AVEC (Tabil *et al.*, 1999). The dryer has overall dimensions of 4318 mm in length and 2235 mm in height. The entire dryer was encased in R10 well-head insulation. It was a semi-closed system, recirculating air in the duct, with an adjustable make up air port and an exhaust air port. A Delhi model 610 fan (Delhi Industries Inc., Delhi, ON) with a ¾ hp Baldor motor (Baldor Electric Co., Ft. Smith, AR) was used to move the air in the duct. The exhaust air port allowed for the expansion of the air, at ambient pressure, as it exited the blower and entered the duct. After passing the exhaust air port, the air was heated by a heating element controlled by an over-temperature sensor. The heated air moved into the upper horizontal section of the dryer and was straightened using a grouping of 76 mm × 914 mm conduit occupying the entire cross-sectional area of the duct. The air coming out of the straightener passed through an orifice plate. The pressure drop across the plate and the air flow rate were measured using flange taps. After turning into the downward, vertical section of the dryer and then turning again toward the lower horizontal portion of the dryer, the air is straightened again using 2.5 mm × 25 mm conduit occupying the entire cross-sectional area of the duct.

Upon leaving the second set of straighteners, the air temperature was measured using an RTD type temperature probe that sent the data to the heater controller. The air then passed over the stainless steel mesh sample tray, which contained the sample to be

dried. The sample tray was connected overhead to an OHAUS Explorer® top loader balance (Ohaus Corp., Florham Park, NJ), where the sample tray was weighed at a predetermined time interval and the weight was recorded directly by the computer. The air then passed through a pair of Type T thermocouples that measured wet and dry bulb temperatures. The wet bulb thermocouple had a moistened wick attached at its junction. The wet and dry bulb temperature data were sampled by a Campbell Scientific CR 21 X data logger (Campbell Scientific Inc., Logan, UT) at a predetermined time interval. The air then passed the make-up air port, which included a damper for throttling the amount of air allowed to enter the dryer. After passing the make-up point, the air entered the fan for recirculation.

2.3.2. EXPERIMENTAL PROCEDURES

2.3.2.1. Sample Preparation

Prior to each drying trial, one bag of a given size of raw *E. angustifolia* roots (approximately 200 g) were removed from the cooler and placed in the laboratory for 10-20 min to equilibrate to room temperature. The roots were then removed from the bag and homogeneously chopped into 1 cm length pieces. The chopped roots were quickly placed into an aluminum plate, which was continuously shaken until the roots were completely mixed. The well-mixed roots were divided into two portions of 90 g each, simultaneously placed into the chambers of preconditioned thin-layer dryer or fluidized-bed dryer. The remaining root samples were used for pre-drying moisture determination.

2.3.2.2. Pre-drying Moisture Determination

The preliminary moisture determination test on Echinacea roots performed by Tabil *et al.* (1999) indicated that oven-drying ground roots at 103°C was better than drying them at 130°C since it would not drive the volatiles off from the roots. It was found that drying at 103°C for 5 h could provide a comparable result as drying at 24 h. This method was used in the present study.

Approximately 20 g of well-mixed and chopped fresh roots were ground using a blender. Three (triplicates) 2-g ground root samples were placed into pre-weighed aluminum moisture dishes. With the dishes slightly open, they were placed in an oven preheated to 103°C. The dishes were removed from the oven 5h later, lids closed, and left in the desiccator until cooled down to room temperature. The dishes were then weighed and the moisture content was calculated on wet basis. The resultant initial moisture was used to calculate the dry-down mass M_f (g) by the following equation:

$$M_f = \frac{M_i(100 - m_i)}{(100 - m_f)} \qquad (6)$$

Where:

M_i = mass of roots before drying, g

m_i = moisture of roots before drying, wet basis percentage

m_f = moisture of roots after dying, wet basis percentage

2.3.2.3. Drying Operation

Laboratory thin-layer dryer and fluidized-bed dryer were preconditioned for at least one hour before the drying operation.

Thin-layer drying process was controlled by a microcomputer system. Before drying was started, the setpoint temperature, time interval for weight monitoring (5 min was selected in this study), and the estimated drying duration were entered through the control panel. Air velocity (3.3 or 4.8 m/s) through the chamber was set by adjusting the fan. The ambient, wet bulb, dry bulb temperature measuring thermocouples and the water reservoir were carefully checked before each drying run. After taring the scale, the Echinacea root samples were placed on the drying tray, the data logger was flagged, and the drying was started. The starting time of drying was recorded along with the initial mass of the sample.

The startup of the fluidized-bed drying was more complex than the thin-layer drying as it was manually controlled. Initially, compressed air control valve was opened widely and the air heating unit was switched on. The air velocity probe was inserted into the port at the top of chamber while the pressure regulator was adjusted until the desired air velocity (3.3 m/s or 4.8 m/s) was achieved. The air pressure shown on the pressure regulator was recorded and used to monitor the stability of air velocity through the chamber. The voltage transformer for the heating temperature control was carefully adjusted until the thermometer at air inlet showed the desired temperature setting. After 30 min preheating, the drying chamber was removed from the system by disconnecting the quick-coupling, leaving the conditioned air running into an insulated PVC chamber, where the wet bulb and dry bulb temperatures were measured by a pair of mercury thermometers. The root samples were loaded into the chamber and the initial weight of

the chamber was measured by a Sartorius® balance (1907 MP8, Sartorius GmbH, Germany), with 5.5 kg capacity and 0.01 g readability. The weighed chamber was then quickly reconnected to the system and the root particles began to be fluidized by the high air velocity. Throughout the drying run, the chamber was removed and weighed every 30 min to generate the moisture loss data. While the chamber was being weighed, running air was introduced to the PVC chamber and the wet bulb and dry bulb temperatures were periodically measured and recorded. PLUS program (Albright, 1990) was used to determine relative humidity based on dry bulb and wet bulb temperatures of the air. Typical thin-layer and fluidized-bed drying process diagrams are presented in Figures 2.3 and 2.4, respectively.

The spice industry prefers to maintain its products at <14% moisture to prevent spoilage by molds and > 5% to avoid crumbling and loss of shape. Therefore, the end point for Echinacea root drying in this study was aimed at 8-10% residual moisture (w.b.). After the dry-down mass was reached, dryers were shut down and the dried roots were quickly removed from the drying chamber and ground into powder with a blender. Three (triplicates) 2-g dried powder roots were sampled for post-drying moisture determination as described below.

2.3.2.4. Post-drying and Equilibrium Moisture Determination

Approximately 2 g of dried roots were weighed into pre-weighed aluminum moisture dishes. Same procedures were then followed as the pre-drying moisture determination described in Section 2.3.2.2.

Considering the perishable condition of the fresh roots, the equilibrium moisture for *E. angustifolia* was determined at the last stage of this experiment, which was completed within a month from the receipt of fresh roots. Each 50 g of small and medium roots were exposed to drying temperatures of 30° and 50°C in both fluidized-bed and thin-layer dryers until the roots stopped losing weight. The dried roots were then ground, and moisture contents were determined following the procedures described above.

2.3.3. DATA ANALYSIS

Page equation (Page, 1949) as described in 1.3.2.2 was used to fit the experimental data. The terms used to evaluate goodness of fit of the tested models are the coefficient of determination, R^2, and the sum of squares of the deviations, SS, between the experimental data and calculated values for the tested model. The R^2, SS, and experimental constants (N, K) were estimated using the SOLVER function of Microsoft Excel 97 Software Package. The equations demonstrating the relationship between drying constants N, K, and drying temperature, air velocity, size of the roots were determined by performing the multiple regression analysis (PROC REG) in SAS (1998).

2.3.4. RESULTS AND DISCUSSION

2.3.4.1. Comparison of Drying Curves

Variations of the drying rate with root moisture content for typical thin-layer and fluidized-bed drying runs are shown in Figures 2.5 and 2.6, respectively. As is evident in these curves and in those of the remaining experimental runs, the constant drying rate

period was absent in the drying of *E. angustifolia* roots. This result is in agreement with the observations of Vaccarezza *et al.* (1974), Mazza and LeMaguer (1980), Kitic and Viollaz (1984), and Rapusas and Driscoll (1995). As explained by Karathanos and Belessiotis (1997), the constant drying rate period is largely associated with the existence of free water and it is usually observed in the drying of inorganic materials (clays, sand, etc.). The absence of constant drying rate period is very common in the drying of food materials and agricultural products, where water is mostly bound. During falling drying rate period, the mechanism of mass transfer is predominately internal, e.g. molecular (liquid) diffusion or vapor diffusion. The internal mass transfer can be enhanced by high temperature, while the effect of air flow rate becomes minimally significant. These hypotheses were strengthened by the observations in this study. Typical drying curves with moisture ratio versus drying time are shown in Figures 2.7-2.15. The effects of drying method, temperature, air velocity, and size of roots on drying rate are best illustrated by these figures. Two sets of drying curves derived from replicate drying trials were presented together under each specific drying condition. There was a good reproducibility of the duplicate drying curves in terms of the final drying times and the moisture content at each point. This suggests that the drying conditions, such as air temperature, air velocity, and root size in duplicate drying trials were consistent and well controlled. Regardless of the drying method used, air temperature was demonstrated as a predominant factor affecting the drying rate of roots. This result is consistent with those observed from drying studies of American ginseng, Kenaf, basil, etc. (Li and Morey, 1987; Muhidong *et al.*, 1992; Rocha *et al.*, 1993). For a given size root dried under the same air velocity, a higher drying temperature gave a higher drying rate with a shorter

drying time. For instance, to achieve the root residual moisture 8-10% (w.b.), the thin-layer drying time for small roots with 0.7 m/s air velocity decreased from 960 min at 30°C to 310 min at 50°C (Fig. 2.7). Similarly, the time for fluidized-bed drying for small roots with 3.3 m/s air velocity decreased from 700 min at 30°C to 270 min at 50°C (Fig. 2.8). Such a large difference in drying time showed that the efficiency of forced-air dryers could be improved by elevating the drying temperature within a practical range. The effect of root size on drying rate in both drying methods was also obvious. The larger the root diameter, the slower the drying rate (Fig. 2.9 and 2.10). It indicates that the roots should be sorted by size prior to drying to avoid over-drying, especially with small roots. This observation may be important to the farmers in relation to the processing of their crops. The effect of air velocity on drying rate in the forced-air thin-layer dryer was not as obvious as the effects of temperature and root size (Fig. 2.11) within the range of velocities studied. This indicates that a higher air velocity does not necessarily give a higher drying rate for *E. angustifolia* roots drying. A similar result was observed with fluidized-bed drying, where fairly high air velocities were applied to effect the fluidization of the roots (Fig. 2.12). Similar conclusions were reached by Simmonds *et al.* (1953) and Abid *et al.* (1990). Simmonds *et al.* (1953) investigated wheat drying in an experimental through-flow dryer, and found that the rate of drying was independent of the air velocities within a range of 1.64-8.74 m/s. The minimal effect of air velocity on drying rate indicates that the drying mechanism of *E. angustifolia* roots might be primarily sub-surface controlled by moisture diffusion, but not externally controlled by the moisture evaporation at the solid surface, where the air flow is critical for removing the surface moisture. Nevertheless, Jayas and Sokhansanj (1989) hypothesized that air

flow could affect the thin-layer drying rate of barley when a low air flow was used. It is possible that in the current study, the range of air velocity applied in the thin-layer drying was not wide enough to show a substantial effect on the drying rate.

Under similar drying conditions (temperature, air velocity, and root size), fluidized-bed drying exhibited a higher drying rate than thin-layer drying. Fluidized-bed drying of small roots at 30°C gave a higher drying rate and shorter drying time when compared to thin-layer drying under similar conditions (Fig 2.13). A similar pattern was observed at 50°C for both small roots and medium roots (Fig. 2.14 and 2.15). This can be attributed to the uniform exposure of roots to a large volume of heated air in fluidized-bed drying, which accelerated both the heat and mass transfer rate between the roots and the surrounding air. Comparable results were reported by Usha *et al.* (1995) on mushroom drying conducted in thin-layer dryer and fluidized-bed dryer. To take a close look of the variations of drying rates in the two drying processes, the models for Figures 2.13 and 2.14 were mathematically differentiated and a pair of new curves with drying rate against drying time are presented in Figures 2.16 and 2.17. The high drying rate in fluidized-bed dryer occurred only for a short of period throughout the entire drying process. The observed initial high drying rate rapidly decreased after the fluidized-bed drying began and reached a low level within 80 and 50 min, at 30° and 50°C, respectively. This low drying rate was maintained during the rest of the fluidized-bed drying process until the desired moisture content (8-10%, w.b.) was achieved. Such a phenomenon became more obvious when the medium roots were dried. This might be due to the fact that extremely fast moisture removal in the initial stages of fluidized-bed

drying considerably shrink the outer layer of the roots, leading to a condition known as case-hardening. The roots in this condition may develop a hard surface, which would obstruct further moisture migration from the inner part to the surface of roots during the later drying stage. Therefore, it can be expected that the bigger the size of roots, the longer it would take for moisture to migrate from inner part to the root surface.

2.3.4.2. Determination of Drying Constants K and N

The equilibrium moisture values of roots under various air conditions are shown in Table 2.1. These values were used in the calculation of drying constants and model fitting. For each specific drying condition reflected by a combination of each level of temperature, air velocity, and root size, data of root moisture ratio derived from replicate drying runs were combined and fitted to Page equation (Page, 1949). The coefficient of determination, R^2, sum of squares of deviation, SS, between experimental data and model predictions, along with the drying constants K and N were calculated using MS Excel. These values are presented in Table 2.2. The Page equation was found to fit the experimental data well for both the thin-layer and fluidized-bed drying trials ($R^2>0.96$). Figures 2.18 and 2.19 show the fitting of Page equation to the arbitrarily chosen sets of experimental data derived from thin-layer drying and fluidized-bed drying. The effects of drying air temperature, air velocity, and root size as well as their combinations on drying constants (K, N) were examined in SAS. The main factors (drying temperature, air velocity and root size) significantly ($p<0.05$) affected both experimental drying constants K and N. A further multiple linear regression analysis indicated that K and N values could best be represented by the following equations:

In thin- layer drying:

$K = 8.05875 \times 10^{-5} T + 0.00414V - 0.0031R + 0.00409$

With a coefficient of determination (R^2) of 0.93, sum of squares (SS) 0.0003 and

$N = 0.00511T - 0.03815V - 0.00612R + 0.77803$

With a coefficient of determination (R^2) of 0.97, sum of squares (SS) 0.0218

In fluidized-bed drying:

$K = 0.00012T + 0.00042V - 0.00837R + 0.04450$

With a coefficient of determination (R^2) of 0.92, sum of squares (SS) 0.0017 and

$N = 0.00482T - 0.00662V - 0.00749R - 0.56584$

With a coefficient of determination (R^2) of 0.92, sum of squares (SS) 0.0206

Where:

T = temperature (°C)

V= air velocity (m/s)

R= root size (0=small roots, 1= medium roots)

2.4. PART B: STUDY OF DRYING EFFECTS ON ECHINACOSIDE RETENTION

2.4.1. MATERIALS AND METHODS

2.4.1.1. Sample Preparation

The root powders obtained from each of the previously described drying treatments were used for echinacoside quantitation by RP-HPLC. To minimize the effect of residual moisture on root samples during analysis, these samples were further freeze-dried for 24 h, vacuum-packed, and stored at –20°C until analysis. As an experimental control, three (triplicates) freeze-dried root samples were ground and prepared for echinacoside content analysis.

2.4.1.2. Moisture Determination

For accurate comparison of the echinacoside content of root samples with varying moisture contents, echinacoside amounts were calculated on dry weight basis after analysis. To determine the dry weight of each root sample prior to echinacoside analysis, a 2-g sub-sample was taken for moisture determination as described in section 2.3.2.4 was followed.

2.4.1.3. Echinacoside Extraction

Duplicate 1 g powdered root samples from each drying treatment were weighed in the 33×80 mm cellulose thimble filters (Whatman International Ltd., Maidstone, England). The samples were then placed on the SOX-TEC system HT2 (1045 Extraction

Unit, Tecater, HÖGANÄS, Sweden) and extracted with 70 mL methanol (HPLC grade) for 1 h (six samples can be extracted simultaneously). After 15 min rinsing, the thimbles were removed from the extractor and the extracts were completely transferred to round bottom flasks. The extracts were evaporated to dryness in a rotary vacuum evaporator at a temperature not higher than 23°C. The residues were re-dissolved first in 6 mL of Milli-Q water, then in 1 mL of methanol, and finally washed with 3 mL of 1:6 (methanol:water) solution.

2.4.1.4. Sep-Pak Cleanup

After preliminary conditioning of Sep-Pak Plus C_{18} cartridges (Waters Co., Miford, MA) with 10 mL methanol (HPLC grade) and 10 mL Milli-Q water, the extracts were passed through the Sep-Pak columns inserted into a vacuum manifold processor (Visiprep DL™, Supelco Inc. Bellefonte, PA). Extracts dissolved in methanol:water (1:6) were put through the Sep-Pak columns, eluted using 10 mL of 1:5, 30 mL of 1:4, and 10 mL of 1:3 methanol:water solutions sequentially. All the eluted fractions were separately collected into test tubes. The fractions eluted by 1:6 and 1:5 methanol:water solution were combined as the first fraction. Fractions eluted by 1:4 and 1:3 methanol:water solution subsequently became the second and third fraction, respectively. This cleanup technique allowed most non-polar compounds in the extracts to be retained by the Sep-Pak cartridge, and all the echinacosides were eluted into the three fractions with highest amount in the second fraction.

2.4.1.5. RP-HPLC Analysis

All three fractions of each extract eluted from the Sep-Pak cartridge were analyzed by the RP-HPLC system. The RP-HPLC analysis was performed using UniPoint HPLC computer system for data handling with JASCO UV-975 intelligent UV/VIS detector operated at 330 nm detection wavelength. Prodigy 150×4.6 mm C_{18} reverse phase column packed with ODS (5 µm) was purchased from Phenomenex (Torance, CA). Samples were taken and injected by a 717 Plus auto-sampler (Waters Co., Miford, MA). Aqueous methanol (water: methanol =1:2.5, v/v) with 1% (v/v) addition of acetic acid (ASC grade) was used as the mobile phase. A flow rate of 1 mL/min and an ambient column temperature were employed. Echinacoside standard was isolated and purified in our laboratory as no commercial echinacoside is available. Throughout the entire analysis procedure, a standard control chart (Fig. 2.20) was used to monitor the concentration of echinacoside in each standard being used in separate analysis runs. The echinacoside amounts in three fractions were added up to obtain the total amount of echinacoside in each extract. The obtained echinacoside amount was converted to percentage on dry weight basis.

2.4.2. DATA ANALYSIS

The echinacoside concentration in each dried root sample was analyzed in duplicate and the mean was reported. The means of treatments were individually compared with the mean of experimental control (echinacoside in freeze-dried sample), and the differences between them were recorded as data for statistical analysis. Four-way

analysis of variance was performed using General Linear Model procedure provided in SAS Statistical Software (SAS, 1998). Comparison of the means was carried out by LSD (Least Significant Difference) test in SAS at $\alpha = 0.05$ significance level.

2.4.3. RESULTS AND DISCUSSION

Figure 2.21 shows typical HPLC chromatograms from echinacoside analysis. The Comparisons of echinacoside retention after thin-layer drying, fluidized-bed drying and freeze-drying are demonstrated in Figures 2.22 and 2.23. When compared to freeze-dried roots, the air-dried roots exhibit some loss of echinacoside. The degree of loss varied largely depending upon the type of dryer and drying conditions applied. The maximum loss of echinacoside could be as high as 73% (30°C, 4.8 m/s air velocity, medium roots, fluidized-bed drying), and the minimum loss could be as little as only 10.4% (50°C, 1.1 m/s air velocity, small roots, thin-layer drying). This result demonstrates that the echinacoside might be quite sensitive to oxygen, which was absent in freeze-drying, but existed throughout the entire air drying procedures. The thin-layer drying was found to be significantly ($P<0.05$) better than the fluidized-bed drying in terms of echinacoside retention. Extremely fast moisture removal at the beginning of fluidized-bed drying can cause dramatic chemical modification, which might offer a possible explanation for the severe echinacoside loss in fluidized-bed drying. No statistically significant difference ($p>0.05$) of echinacoside loss was found between 30°C and 50°C drying. This result reveals that 50°C drying with much higher drying rate is acceptable from the view of

overall drying efficiency and echinacoside preservation. The experimental data showed that the effect of two levels of air velocity on echinacoside retention was insignificant ($p>0.05$). Considering the minor effect of air velocity on drying rate, therefore, a lower air velocity should be preferred to avoid the detrimental oxidation of echinacoside while meeting the need of energy conservation. It should be noted that the size of root was observed to be an important factor influencing the echinacoside retention in dehydrated roots. Highly significant ($P<0.05$) difference of echinacoside loss was observed between dried small and dried medium roots. Dried small roots were found to have higher echinacoside retention as compared to that of dried medium roots. The higher retention of echinacoside in dried small roots might be attributed to the faster drying rate, and hence a shorter drying duration in either thin-layer dryer or fluidized-bed dryer.

In an attempt to prove the relationship between drying rate and echinacoside retention, a liner regression was performed with the drying constants K, which characterizes the rate of moisture removal from the roots, versus the relative echinacoside values in different treatments. The drying constant K in each individual drying trial was calculated and utilized to correlate with echinacoside concentration from each individual HPLC analysis. The values used in the regression analysis are presented in Appendix I. The relationship between K values and echinacoside loss was not significant in both thin-layer drying and fluidized-bed drying, with regression coefficient (R^2) of 0.625 and 0.172, respectively. Even though the regression analysis was inconclusive, the lack of significant regression coefficient was possibly due to the uncontrolled relative humidity of air in the dryer, or the variance in initial moisture among the roots being dried on

separate days. Both these factors can lead to a comparatively large variance of drying constant K even in the duplicate drying trials.

2.5. CONCLUSION

The air drying of *E. angustifolia* roots occurred through a falling drying rate stage, where the mass transfer was internally controlled possibly by moisture diffusion. Consequently, the air velocity, which is influential on moisture removal in constant drying rate period, has little effect on drying rate of *E. angustifolia* roots. The air temperature and size of roots are important factors affecting the drying rates of roots in thin-layer drying and fluidized-bed drying. The higher the temperature or the smaller the root size, the faster the drying rate. Therefore, the efficiency improvement of forced-air dryers for *E. angustifolia* roots can be expected by increasing the drying temperature within a practical range. Also, it will be advisable to separately dry the small roots and medium roots so as to avoid over-drying of small roots as seen when the small and medium roots are mixed. The Page equation (Page, 1949) was found to nicely fit the experimental data derived from both drying methods. By correlating the experimental drying constants K and N to drying temperature, air velocity, and size of roots, the duration of Echinacea root drying can be predicted in both drying methods tested.

Under similar drying conditions, fluidized-bed drying provided a higher drying rate than thin-layer drying at the initial drying stage. The high drying rate in fluidized-bed dropped rapidly within a short period of time, and a low drying rate was observed during

the rest of drying procedure. The case-hardening of roots may have contributed to this undesirable case. Moreover, it was found that the echinacoside retention in fluidized-bed dried roots was poor. Therefore, thin-layer dryer might be more suitable than fluidized-bed dryer for *E. angustifolia* roots dehydration, with respect to overall drying efficiency and the medicinal quality of dehydrated roots. Echinacoside loss was not affected ($p>0.05$) by temperature or air velocity in the two drying methods. Based on these results, it can be concluded that thin-layer drying with drying temperature of 50°C and air velocity of 0.7 m/s would be the optimum choice for *E. angustifolia* roots dehydration under our experimental conditions.

2.6. REFERENCES

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Table 2.1. Equilibrium moisture (EM) content of Echinacea roots

Drying Method	Setpoint Temperature (°C)	Drying Conditions	EM Content (% d.b.)
Fluidized-bed	50	T_{db}: 50°C T_{wb}: 19.5 °C Relative humidity: 2.3% Time: 16 h Root size: small	4.03
Fluidized-bed	50	T_{db}: 50°C T_{wb}: 18.5°C Relative humidity: 0.7% Time: 27 h Root size: medium	4.56
Fluidized-bed	30	T_{db}: 30°C T_{wb}: 13.5°C Relative humidity: 11.2% Time:40 h Root size: small	6.37
Fluidized-bed	30	T_{db}: 30°C T_{wb}: 13.5°C Relative humidity: 11.2% Time: 40 h Root size: medium	7.48
Thin-layer	50	T_{db}: 49.2°C T_{wb}: 21.2°C Relative humidity: 5.9% Time: 33 h Root size: small	4.20
Thin-layer	50	T_{db}: 49.2°C T_{wb}: 21.2°C Relative humidity: 5.9% Time: 33 h Root size: medium	4.33
Thin-layer	30	T_{db}: 29.6°C T_{wb}: 14.1°C Relative humidity: 14.5% Time: 46 h Root size: small	6.64
Thin-layer	30	T_{db}: 29.6°C T_{wb}: 14.1°C Relative humidity: 14.5% Time: 46h Root size: medium	7.60

Table 2.2. Drying constants in Page model (1949) of Echinacea roots at various conditions

Drying Regime	Drying constants of the Model $MR = e^{-Kt^N}$
I. Fluidized-bed drying	
Sample Code: 1FL5033S[1]	
T_{db}: 50 °C	
T_{wb}: 19.5°C	
RH: 2.3%	K= 0.05181
Air Velocity: 3.3 m/s	N= 0.79629
Root size: Small	
Drying time: 270 min	
Sample Code: 2FL5033S	R^2= 0.99
T_{db}: 50 °C	SS= 0.0061
T_{wb}: 18.7°C	
RH: 1.0%	
Air Velocity: 3.3 m/s	
Root size: Small	
Drying time: 240 min	
Sample Code: 2FL5048S	
T_{db}: 50°C	
T_{wb}: 17.5°C	
RH: N/D	K= 0.05282
Air Velocity: 4.8 m/s	N= 0.07671
Root size: Small	
Drying time: 270 min	
Sample Code: 3FL5048S	R^2= 0.97
Tdb: 50°C	SS= 0.0046
Twb: 19.4°C	
RH: 2.2%	
Air Velocity: 4.8 m/s	
Root size: Small	
Drying time: 270 min	

Sample Code:	**1FL5033M**	
T_{db}:	50°C	
T_{wb}:	18.5°C	
RH:	0.7%	K= 0.042406
Air Velocity:	3.3 m/s	N= 0.789656
Root size:	Medium	
Drying time:	420 min	R^2= 0.98
Sample Code:	**2FL5033M**	SS= 0.0018
T_{db}:	50°C	
T_{wb}:	19.8°C	
RH:	2.8%	
Air Velocity:	3.3 m/s	
Root size:	Medium	
Drying time:	390 min	
Sample Code:	**2FL5048M**	
T_{db}:	50°C	
T_{wb}:	18.5°C	
RH:	0.7%	K= 0.045291
Air Velocity:	4.8 m/s	N= 0.753023
Root size:	Medium	
Drying time:	450 min	R^2= 0.99
Sample Code:	**3FL5048M**	SS = 0.0069
T_{db}:	50°C	
T_{wb}:	20.1°C	
RH:	3.3%	
Air Velocity:	4.8 m/s	
Root size:	Medium	
Drying time:	450 min	
Sample Code:	**1FL3033S**	
T_{db}:	30°C	
T_{wb}:	13.5°C	
RH:	11.2%	K=0.051499
Air Velocity:	3.3 m/s	N=0.664389
Root size:	Small	
Drying time:	660 min	R^2= 0.97
Sample Code:	**2FL3033S**	SS= 0.0187
T_{db}:	30°C	
T_{wb}:	13.2°C	
RH:	10%	
Air Velocity:	3.3 m/s	
Root size:	Small	
Drying time:	690 min	

Sample Code:	**1FL3048S**	
T_{db}:	30°C	
T_{wb}:	13.8°C	
RH:	12.4%	K=0.047912
Air Velocity:	4.8 m/s	N= 0.701531
Root size:	Small	
Drying time:	630 min	R^2= 0.98
Sample Code:	**2FL3048S**	SS= 0.0272
T_{db}:	30°C	
T_{wb}:	13.5°C	
RH:	11.2%	
Air Velocity:	4.8 m/s	
Root size:	Small	
Drying time:	630 min	
Sample Code:	**1FL3033M**	
T_{db}:	30°C	
T_{wb}:	12.5°C	
RH:	7.3%	K=0.040205
Air Velocity:	3.3 m/s	N=0.691861
Root size:	Medium	
Drying time:	660 min	R^2= 0.99
Sample Code:	**2FL3033M**	SS= 0.0165
T_{db}:	30°C	
T_{wb}:	13.8°C	
RH:	12.4%	
Air Velocity:	3.3 m/s	
Root size:	Medium	
Drying time:	690 min	
Sample Code:	**2FL3048M**	
T_{db}:	30°C	
T_{wb}:	14.2°C	
RH:	13.9%	K= 0.042732
Air Velocity:	4.8 m/s	N= 0.671568
Root size:	Medium	
Drying time:	900 min	R^2= 0.98
Sample Code:	**3FL3048M**	SS=0.0198
T_{db}:	30°C	
T_{wb}:	13.6°C	
RH:	11.6%	
Air Velocity:	4.8 m/s	
Root size:	Medium	
Drying time:	870 min	

Drying Regime		Drying constants of the Model $MR = e^{-Kt^N}$
II. Forced-air thin-layer drying		
Sample Code: 1T5011S		
T_{db}:	49.28°C	
T_{wb}:	21.48°C	
RH:	6.26%	K= 0.014146
Air Velocity:	1.1 m/s	N= 0.981778
Root size:	Small	
Drying time:	400 min	R^2= 0.99
Sample Code: 2T5011S		SS= 0.0648
T_{db}:	49.41°C	
T_{wb}:	21.24°C	
RH:	5.71%	
Air Velocity:	1.1 m/s	
Root size:	Small	
Drying time:	355 min	
Sample Code: 1T5007S		
T_{db}:	49.13°C	
T_{wb}:	21.61°C	
RH:	6.63%	K= 0.011023
Air Velocity:	1.1m/s	N= 0.999013
Root size:	Small	
Drying time:	310 min	R^2= 0.99
Sample Code: 2T5007S		SS= 0.0149
T_{db}:	49.02°C	
T_{wb}:	21.87°C	
RH:	7.23.%	
Air Velocity:	1.1m/s	
Root size:	Small	
Drying time:	295 min	

Sample Code: 1T5011M		
T_{db}:	49.24°C	
T_{wb}:	21.69°C	
RH:	6.64%	K= 0.008783
Air Velocity:	1.1m/s	N= 0.978761
Root size:	Medium	
Drying time:	670 min	R^2= 0.99
Sample Code: 2T5011M		SS= 0.1255
T_{db}:	49.44°C	
T_{wb}:	23.64°C	
RH:	10.34%	
Air Velocity:	1.1m/s	
Root size:	Medium	
Drying time:	735 min	

Sample Code: 1T5007M		
T_{db}:	49.34°C	
T_{wb}:	21.86°C	
RH:	6.94 %	K= 0.008316
Air Velocity:	0.7 m/s	N= 1.003132
Root size:	Medium	
Drying time:	550 min	R^2 = 0.99
Sample Code: 2T5007M		SS = 0.0257
T_{db}:	49.32°C	
T_{wb}:	21.87°C	
RH:	6.94 %	
Air Velocity:	0.7 m/s	
Root size:	Medium	
Drying time:	530 min	

Sample Code: 1T3011S		
T_{db}:	29.63°C	
T_{wb}:	13.67°C	
RH:	11.85%	K= 0.010695
Air Velocity:	1.1 m/s	N= 0.889076
Root size:	Small	
Drying time:	720 min	R^2= 0.99
Sample Code: 2T3011S		SS= 0.0207
T_{db}:	29.63°C	
T_{wb}:	16.22°C	
RH:	12.99%	
Air Velocity:	1.1 m/s	
Root size:	Small	
Drying time:	730 min	

Sample Code: 1T3007S		
T_{db}:	29.56°C	
T_{wb}:	14.26°C	
RH:	15.39%	K= 0.009359
Air Velocity:	0.7m/s	N= 0.900687
Root size:	Small	
Drying time:	960	R^2= 0.99
Sample Code:2T3007S		SS= 0.0611
T_{db}:	29.58°C	
T_{wb}:	14.79°C	
RH:	17.21%	
Air Velocity:	0.7m/s	
Root size:	Small	
Drying time:	750	
Sample Code:1 T3011M		
T_{db}:	29.67°C	
T_{wb}:	13.36°C	
RH:	11.54%	K= 0.008754
Air Velocity:	1.1 m/s	N= 0.878204
Root size:	Medium	
Drying time:	1454 min	R^2= 0.99
Sample Code: 2T3011M		SS= 0.0397
T_{db}:	29.73°C	
T_{wb}:	14.10°C	
RH:	14.81%	
Air Velocity:	1.1 m/s	
Root size:	Medium	
Drying time:	1350 min	
Sample Code: 1T3007M		
T_{db}:	29.64°C	
T_{wb}:	14.73°C	
RH:	16.90 %	K= 0.005904
Air Velocity:	0.7 m/s	N= 0.907660
Root size:	Medium	
Drying time:	1560 min	R^2= 0.99
Sample Code: 2T3007M		SS= 0.2723
T_{db}:	29.66°C	
T_{wb}:	14.21°C	
RH:	14.65 %	
Air Velocity:	0.7 m/s	
Root size:	Medium	
Drying time:	1370 min	

[1]Explanation of symbols (Appendix II)

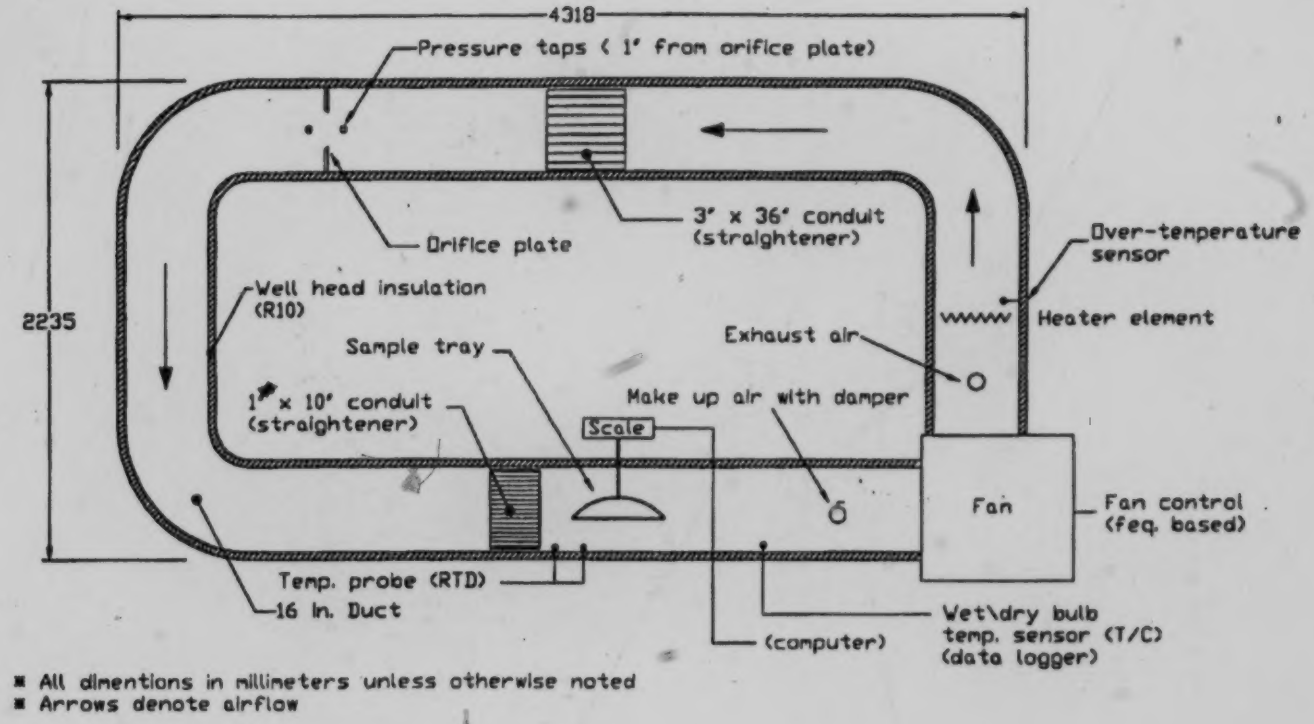
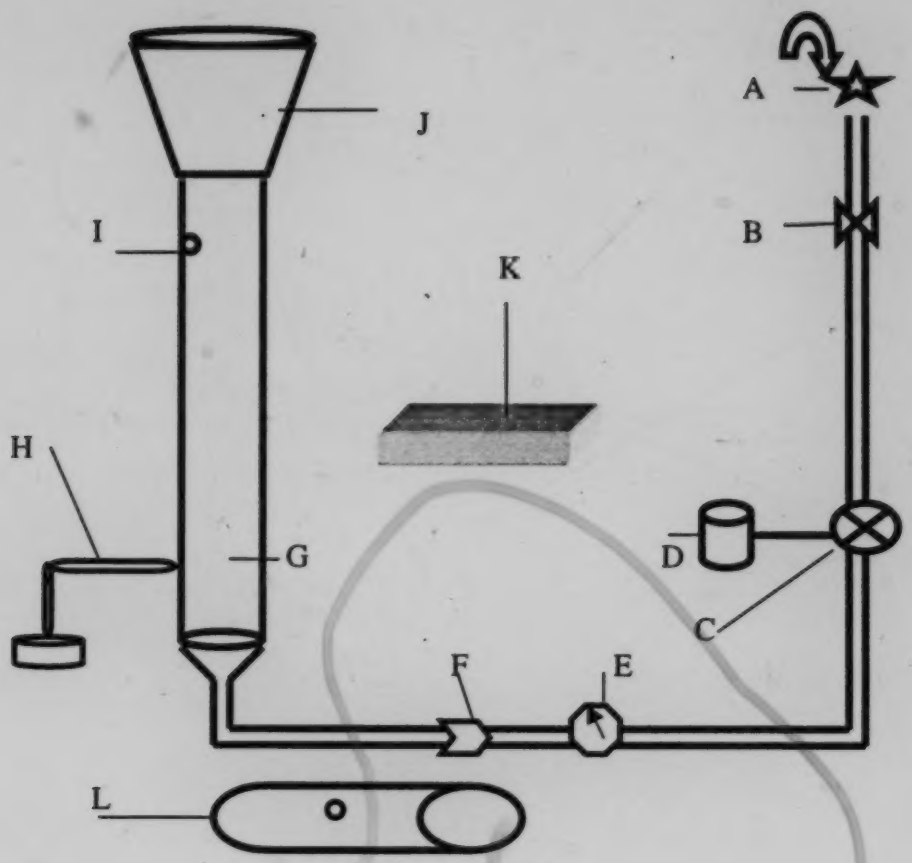


Figure 2.1. Schematic diagram of the thin-layer dryer Tabil *et al* (1999)



A: compressed air control valve
B: gas regulator
C: heating unit
D: voltage transformer
E: thermometer
F: quick coupling
G: drying chamber
H: knocking device
I: air velocity measurement port
J: air velocity reducer
K: scale
L: PVC chamber for dry & wet bulb temperature measurement

Figure. 2.2. Schematic diagram of fluidized-bed dryer

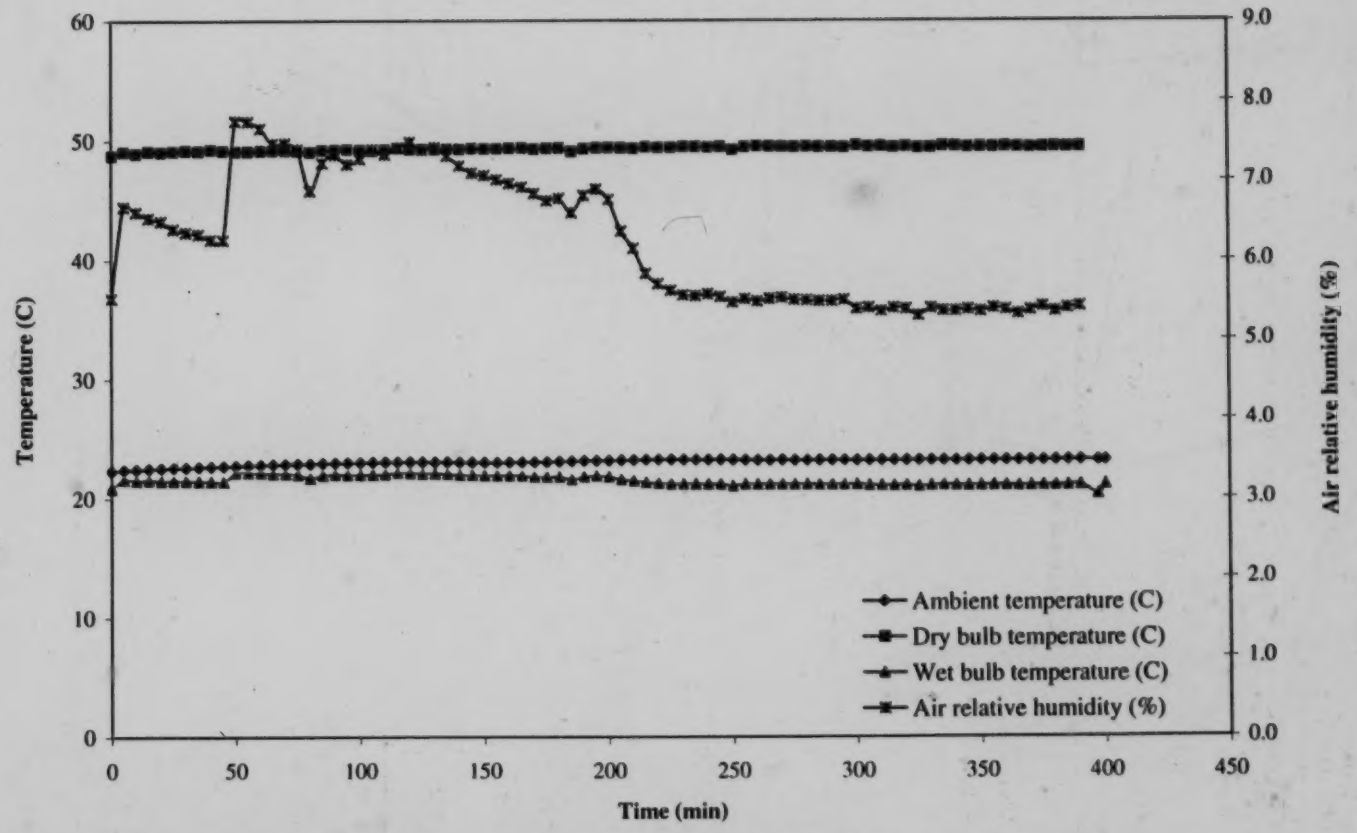


Figure 2.3. Operation history of a typical thin-layer drying process (T=50C, V=1.1 m/s, small roots)

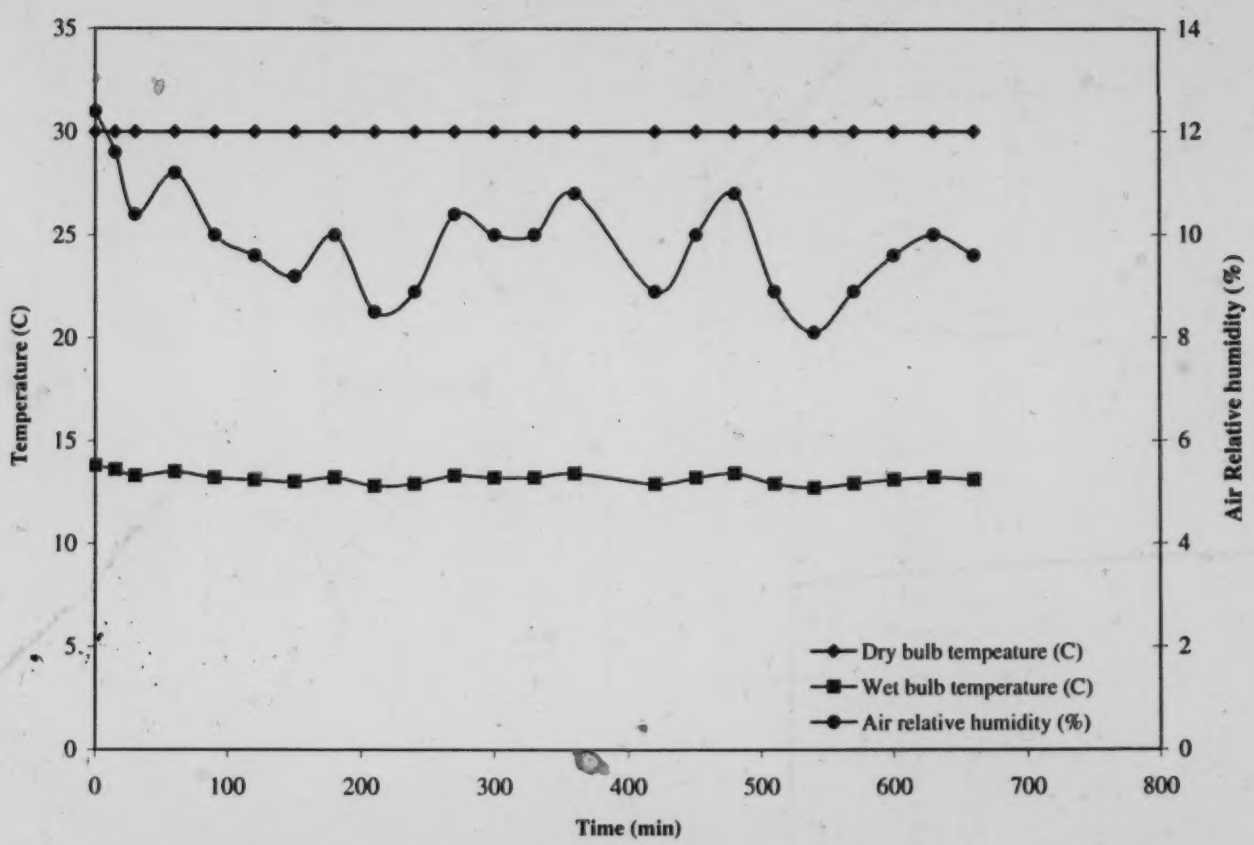


Figure 2.4. Operation history of a typical fluidized-bed drying process (T=30C, V=3.3 m/s, small roots)

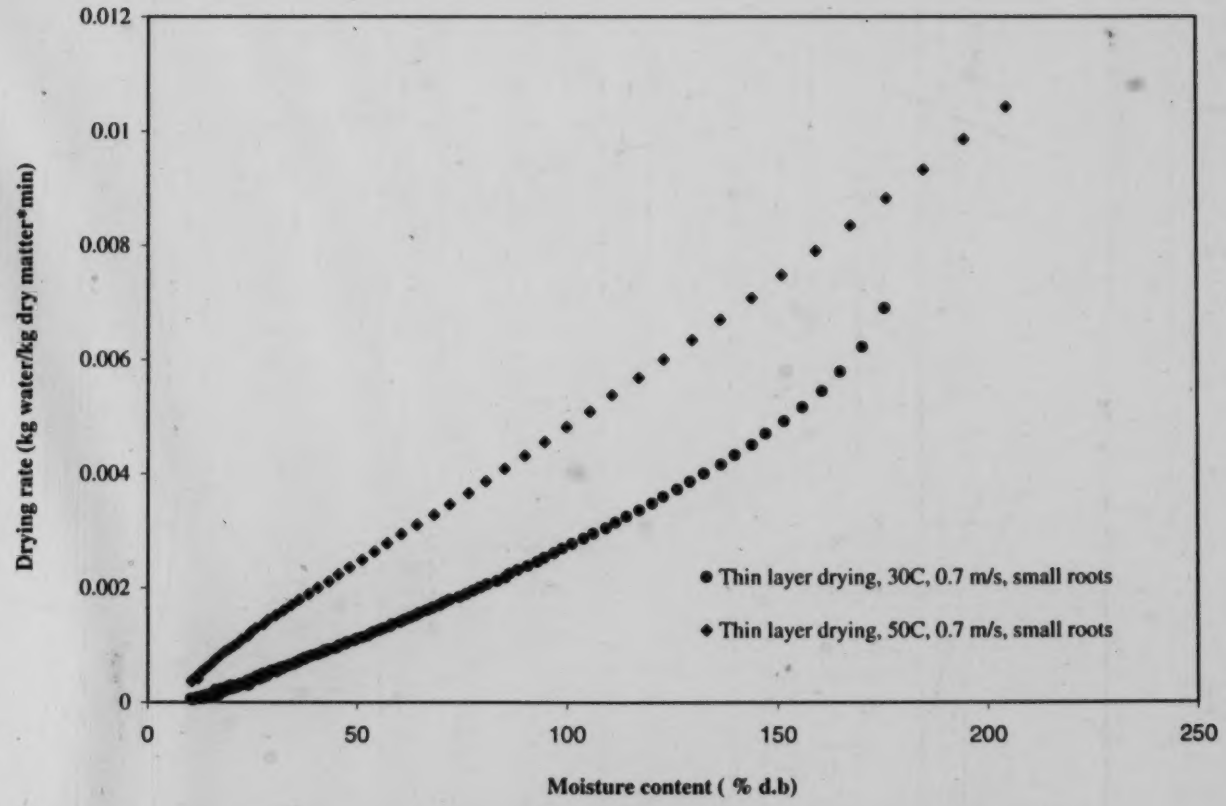


Figure 2.5. Drying rate variation with root moisture during thin-layer drying

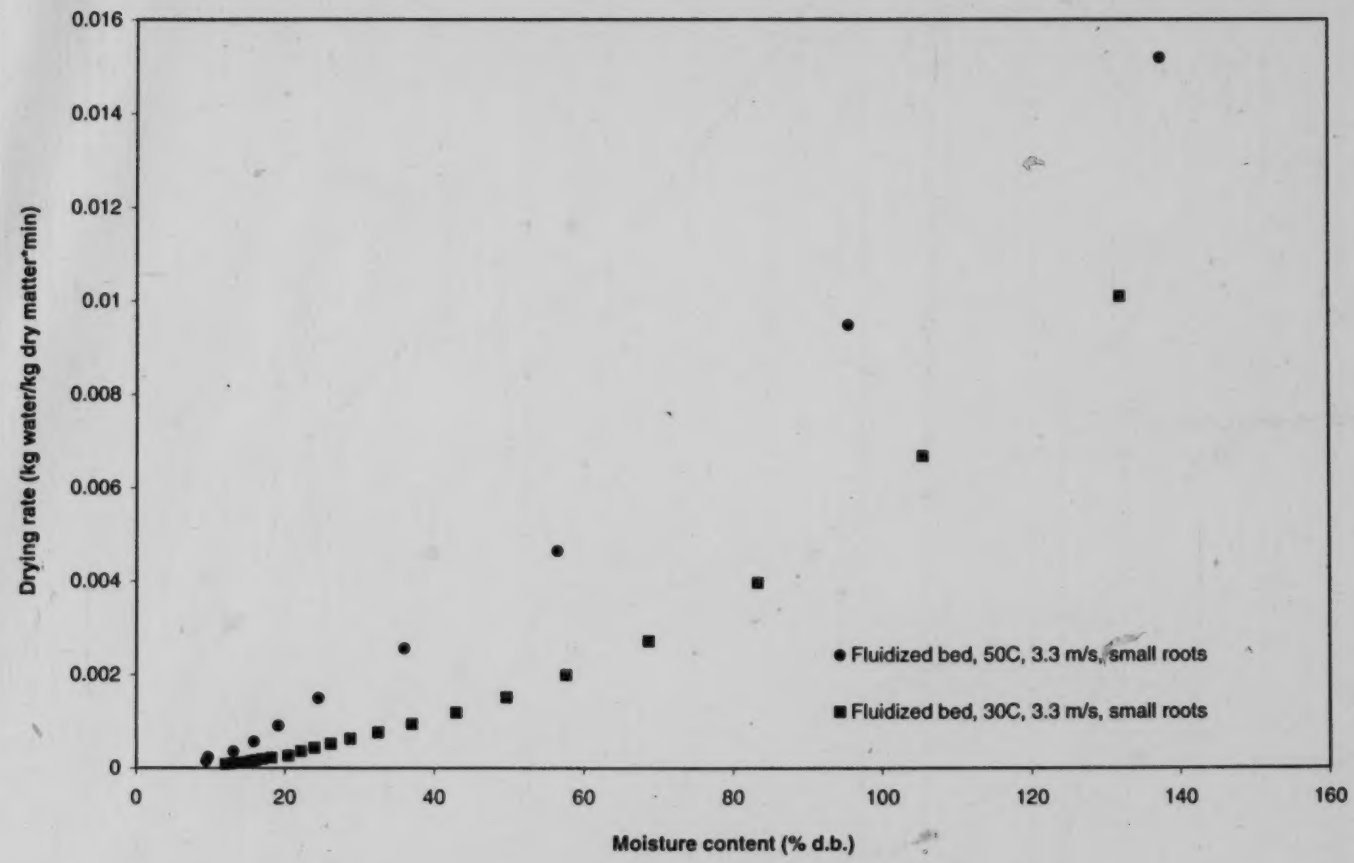


Figure 2.6. Drying rate variation with root moisture during fluidized-bed drying

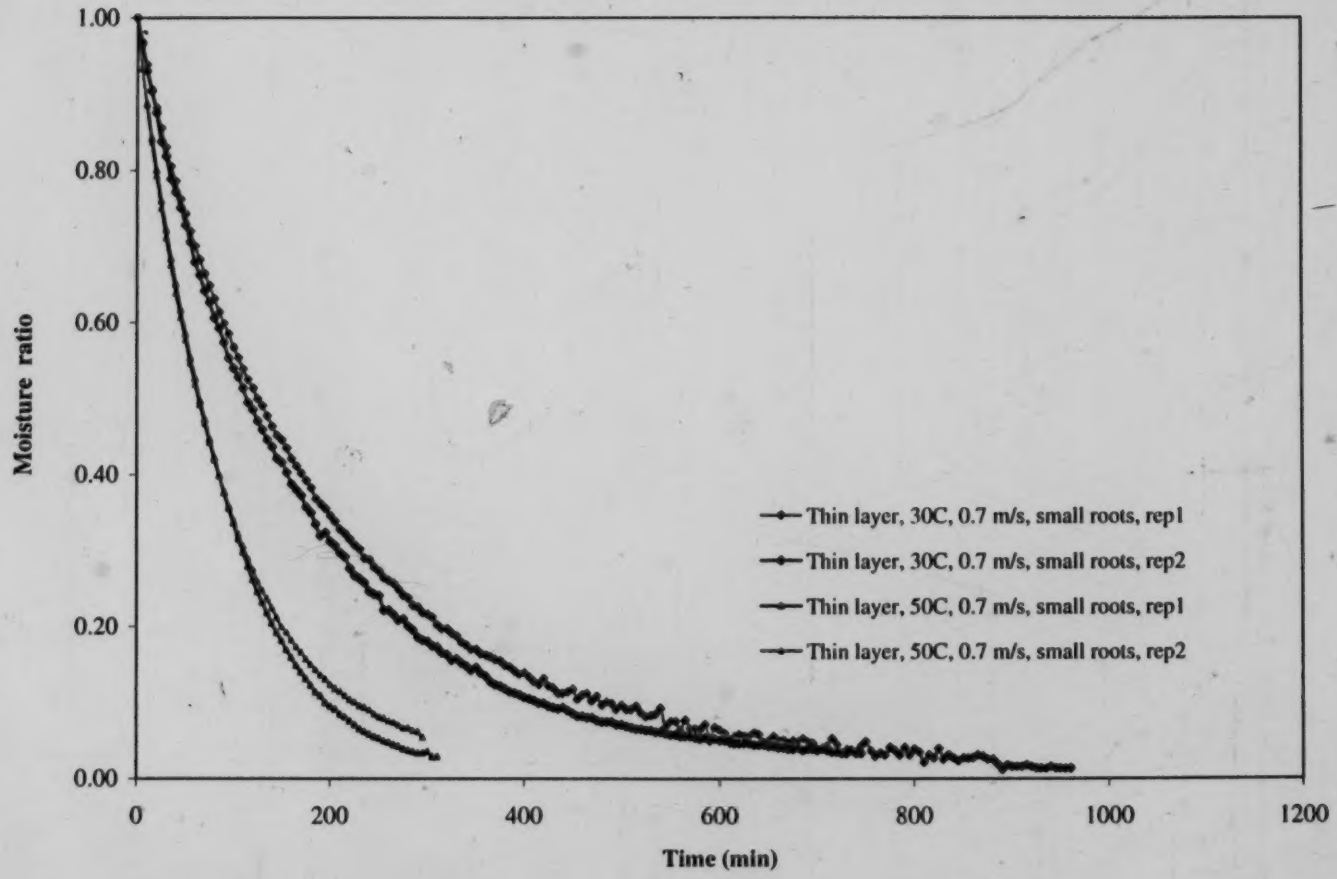


Figure 2.7. Effect of temperature on moisture change during thin-layer drying of root

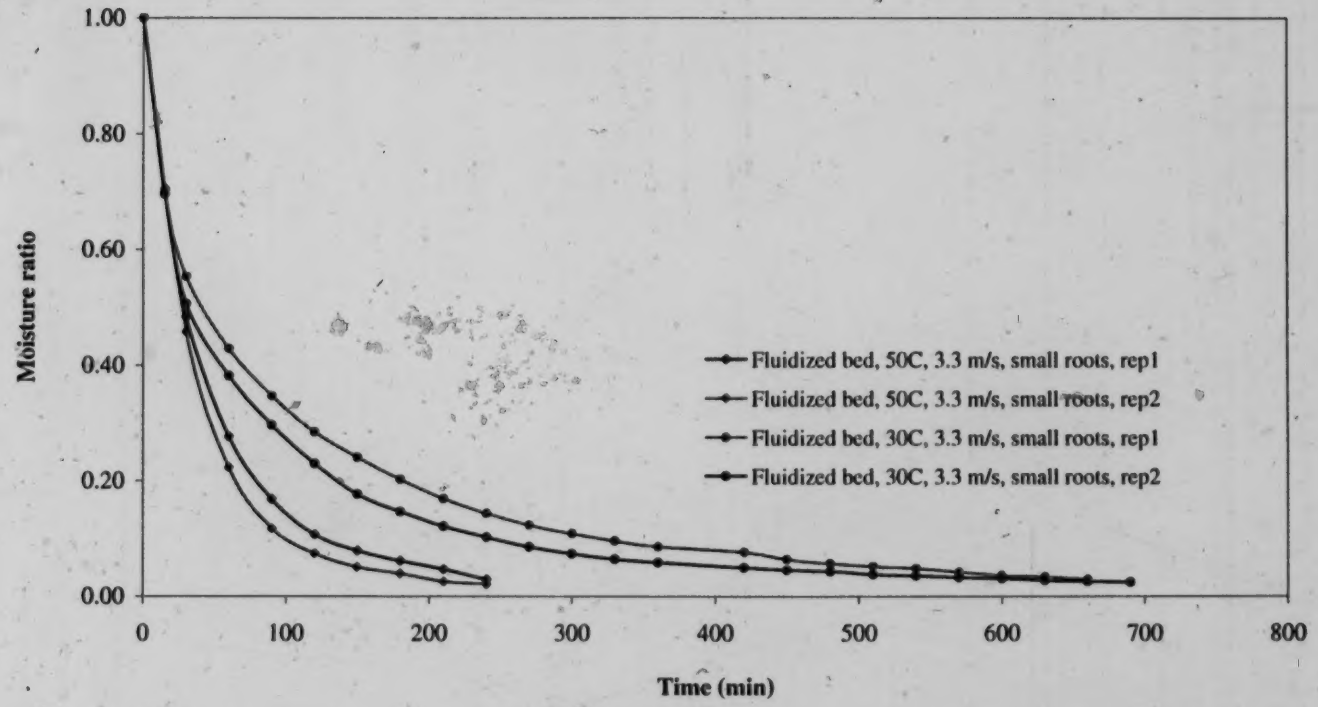


Figure 2.8. Effect of temperature on moisture change during fluidized-bed drying of root

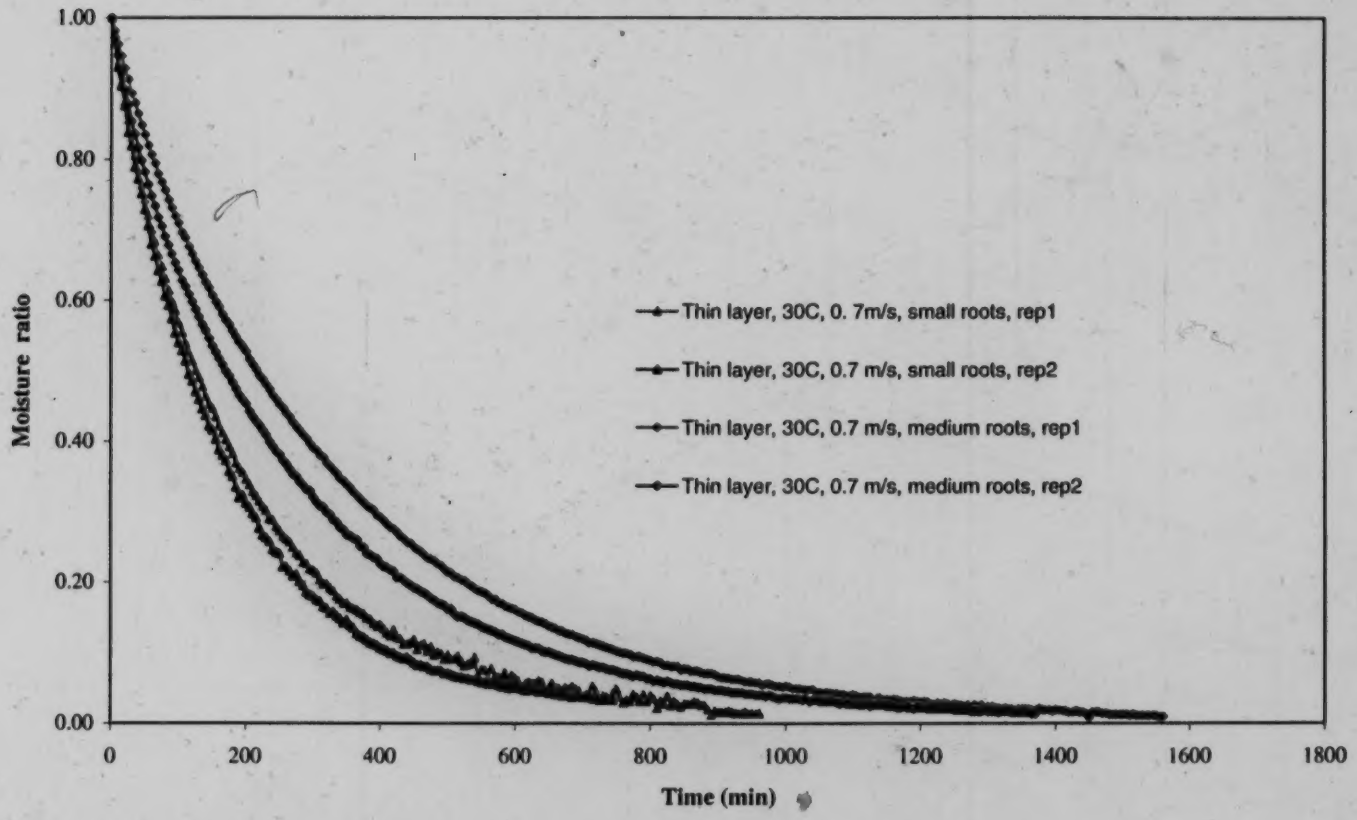


Figure 2.9. Effect of root size on moisture change during thin-layer drying of root

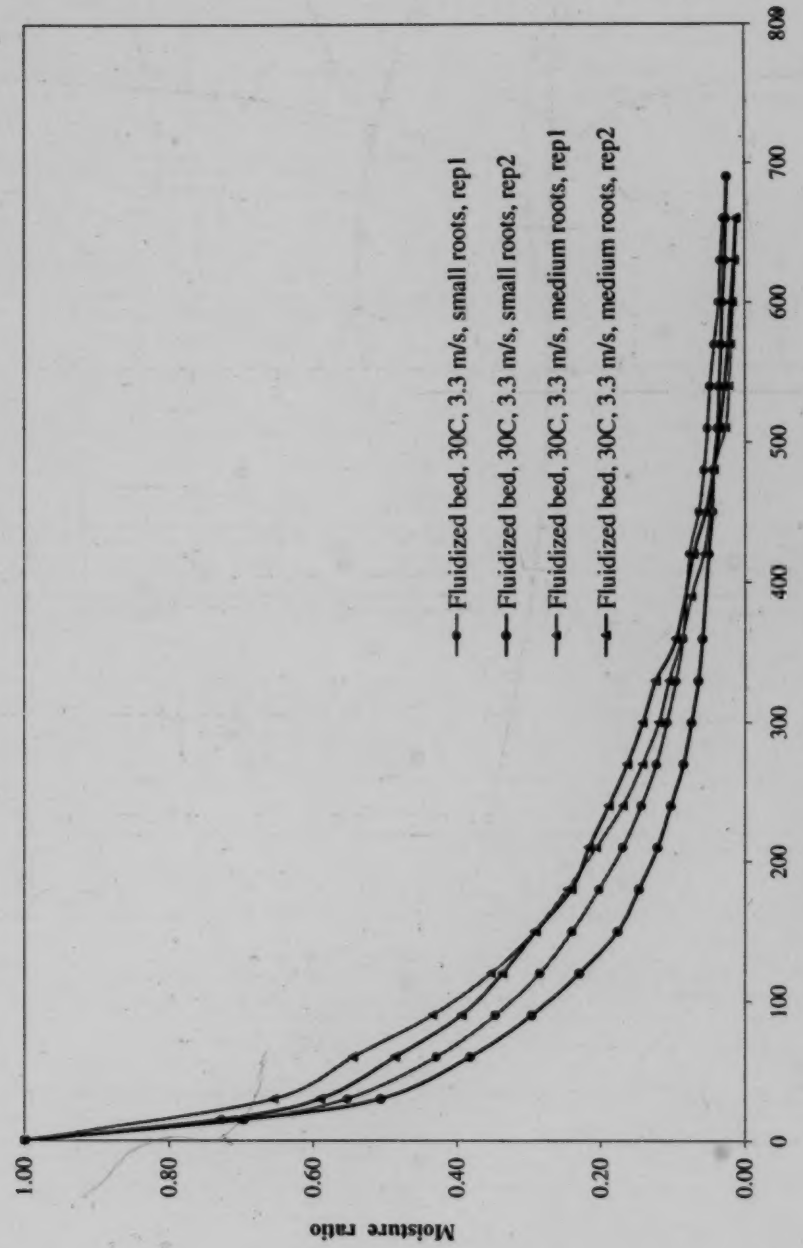


Figure 2.10. Effect of root size on moisture change during fluidized-bed drying of root

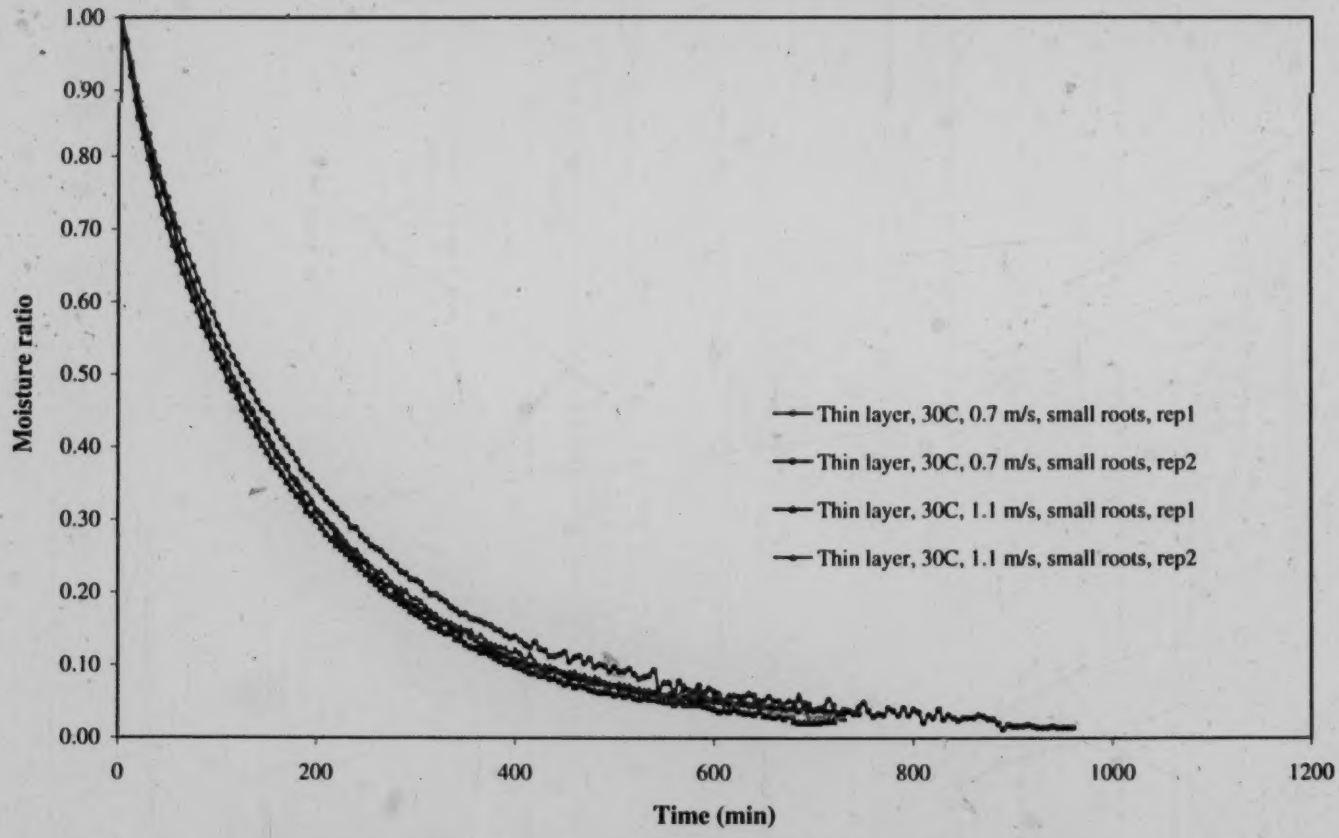


Figure 2.11. Effect of air velocity on moisture change during thin-layer drying of root

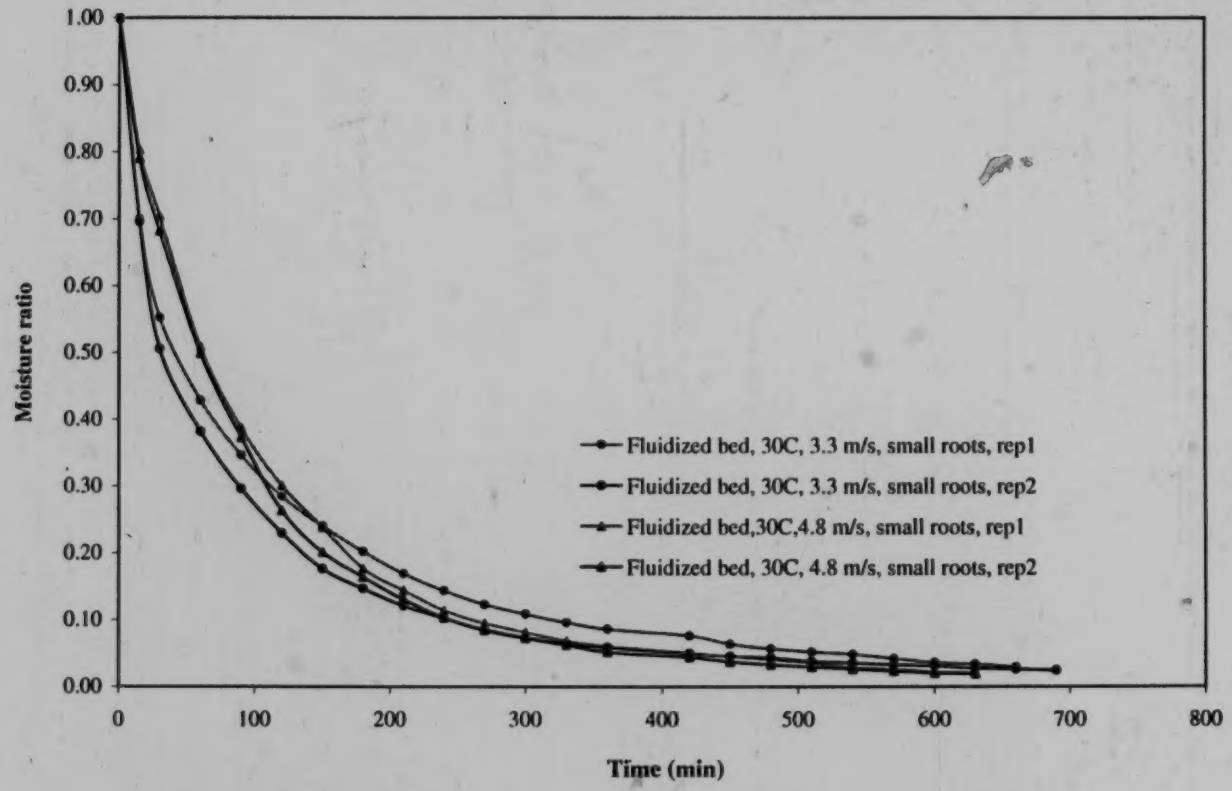


Figure 2.12. Effect of air velocity on moisture change during fluidized-bed drying of root

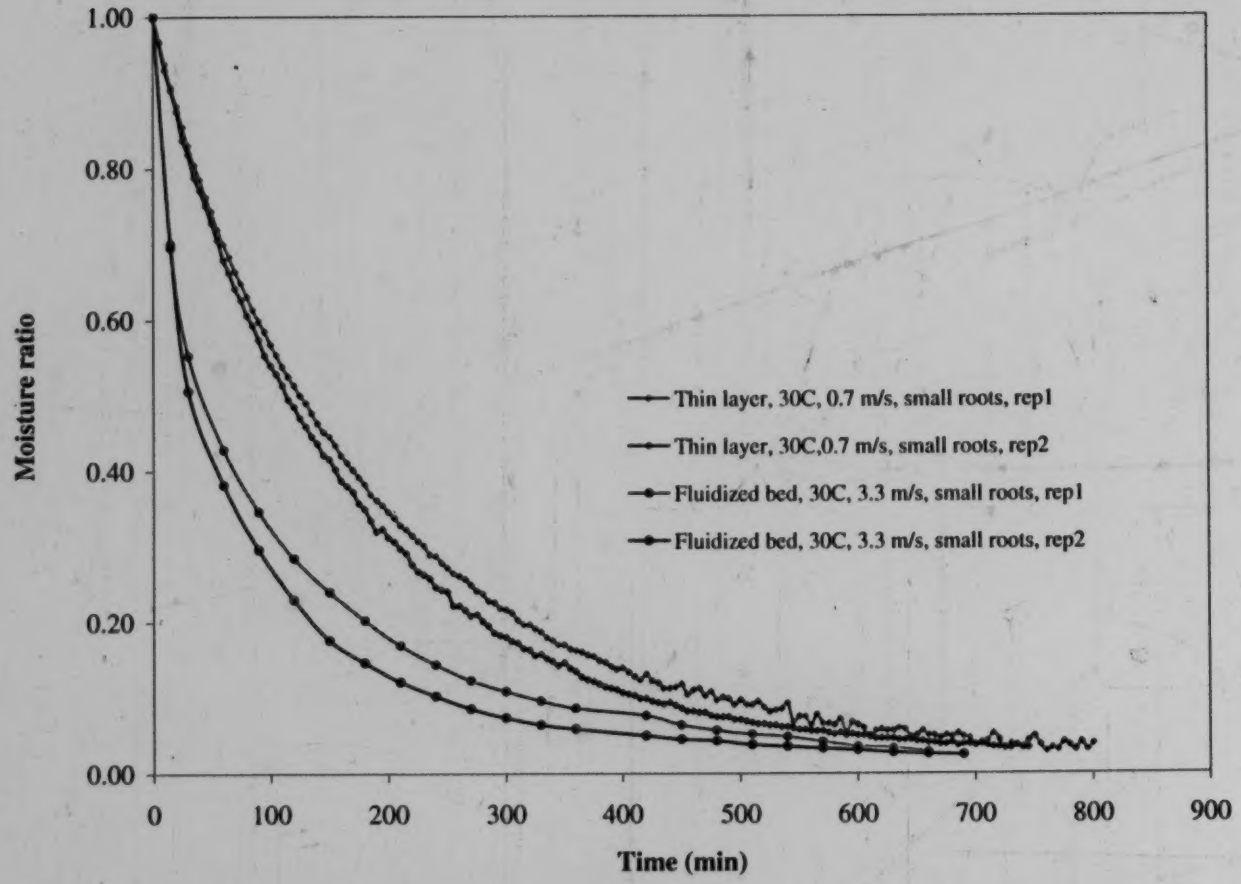


Figure 2.13. Comparison of moisture change in two drying methods (T=30C, small roots)

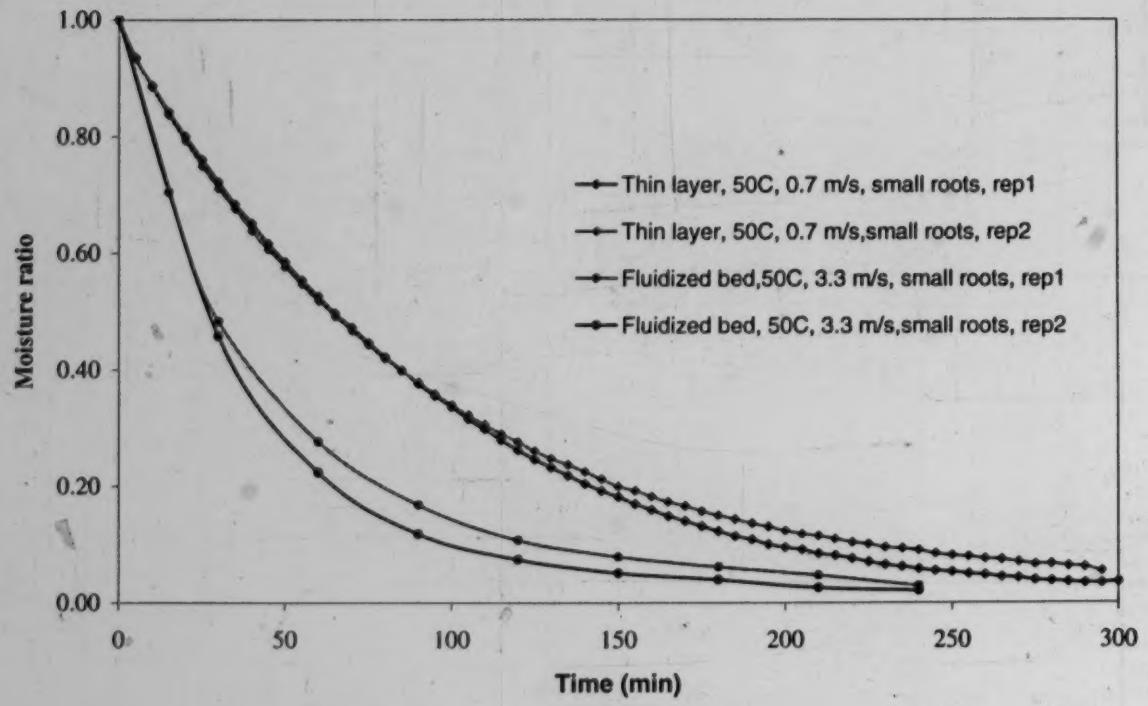


Figure 2.14. Comparison of moisture change in two drying methods (t=50C, small roots)

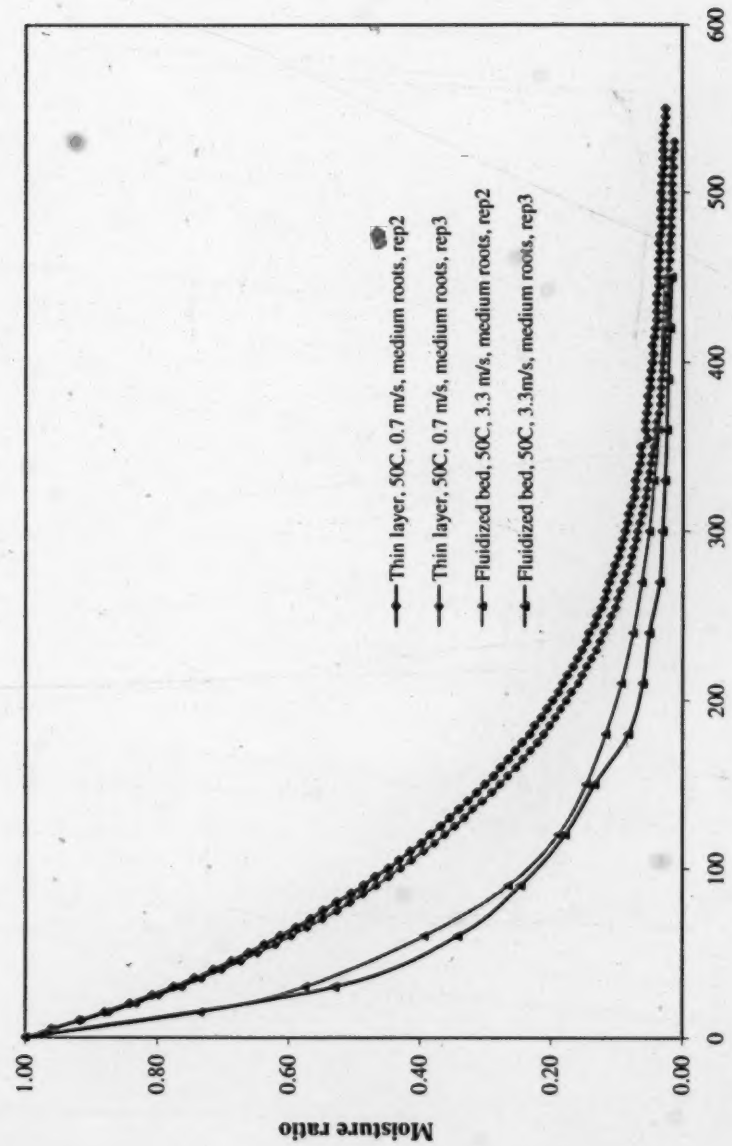


Figure 2.15. Comparison of root moisture change in two drying methods (T=50C, medium roots)

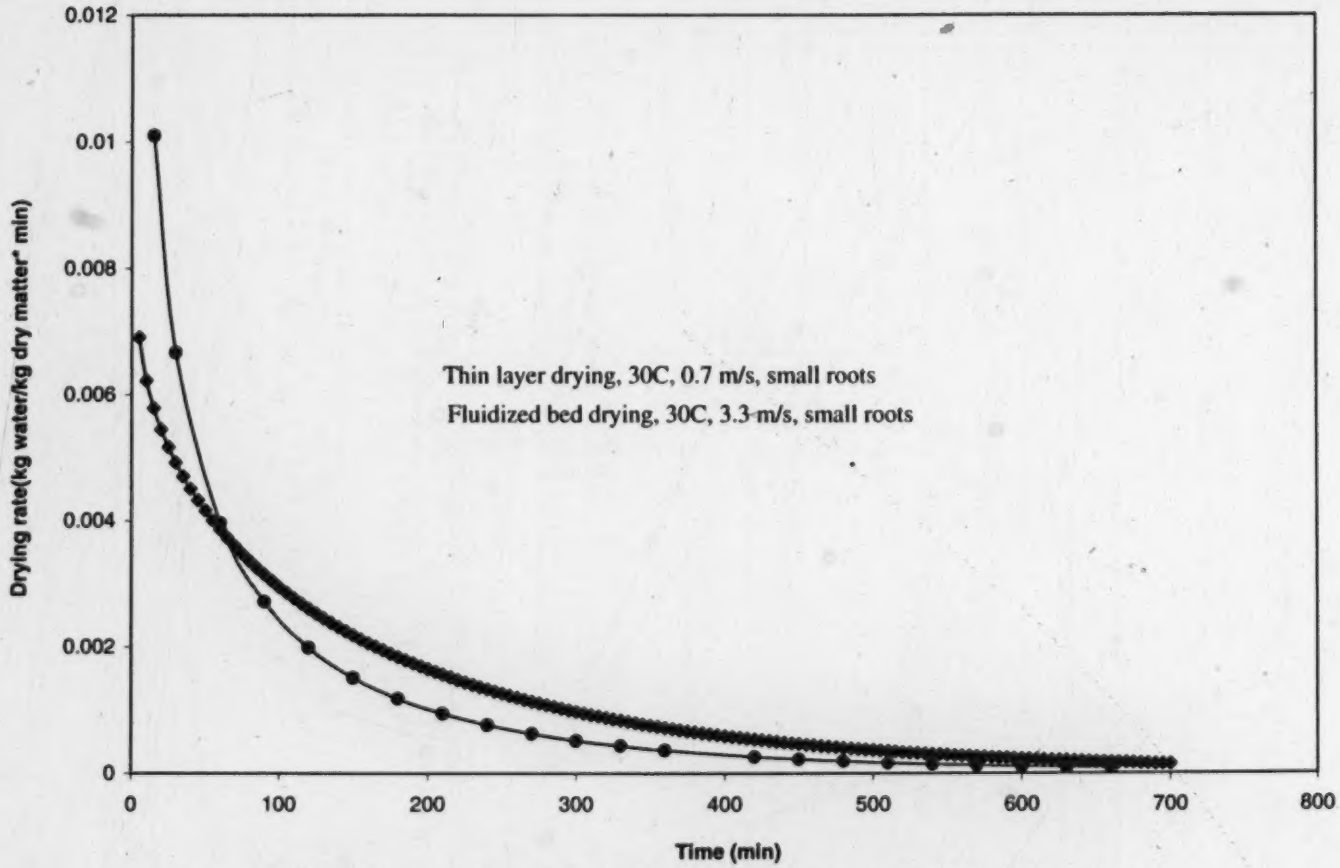


Figure 2.16. Drying rate change with time in two drying methods (T=30C, small roots)

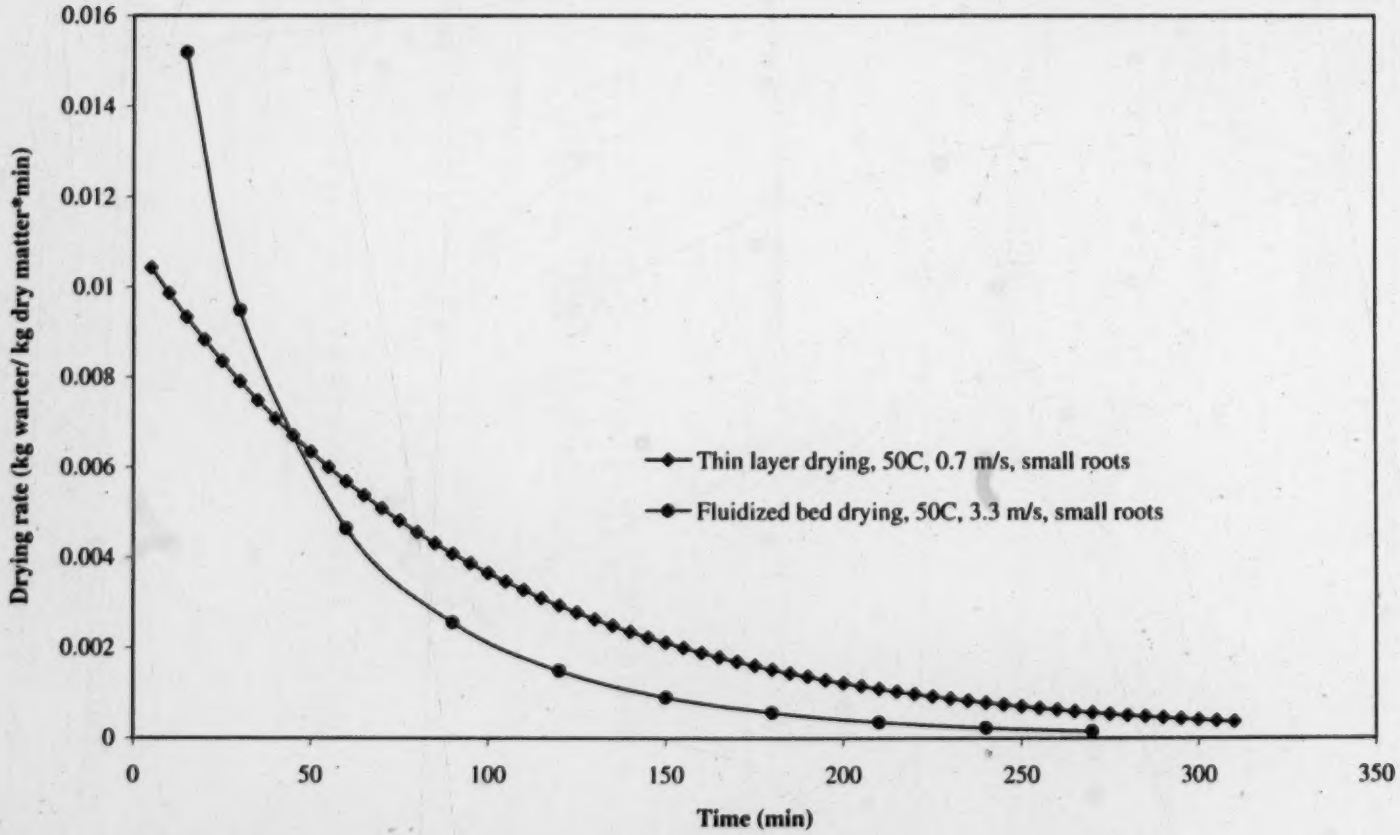


Figure 2.17. Drying rate change with time in two drying methods (T=50C, small roots)

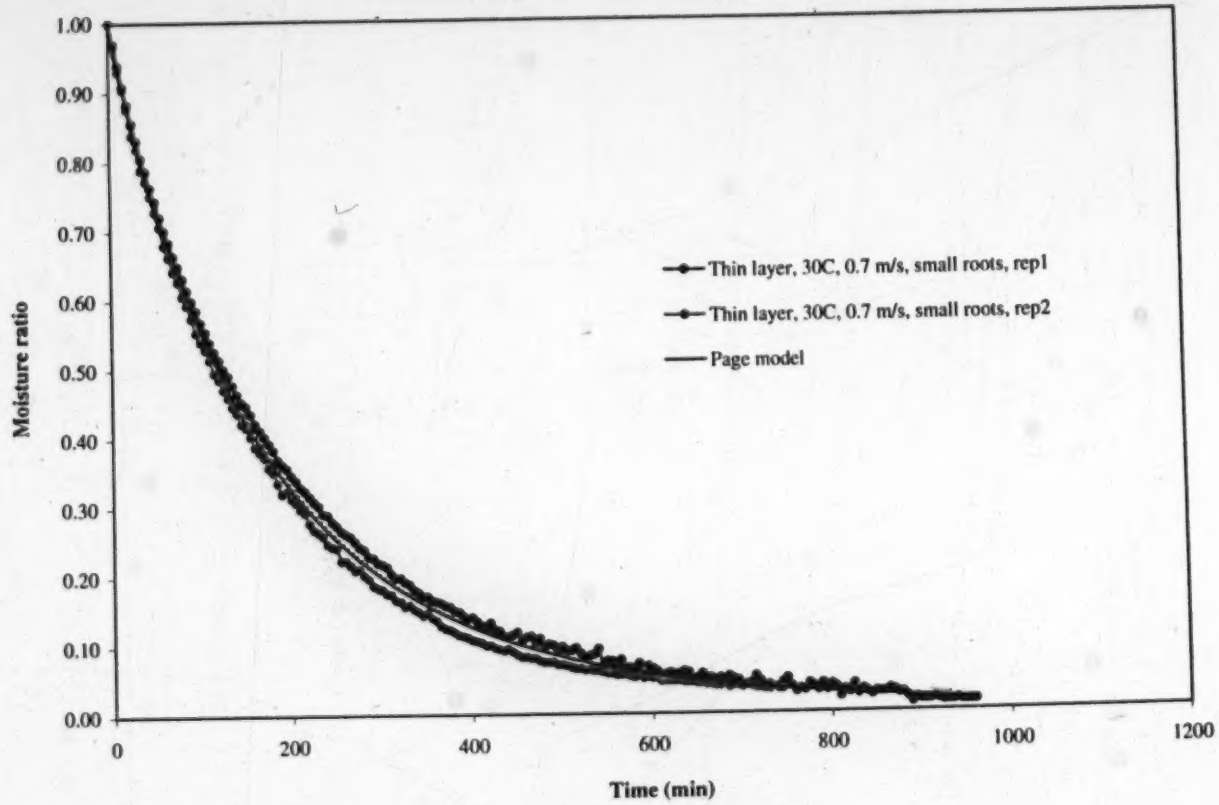


Figure 2.18. Fitting Page model to thin-layer drying data

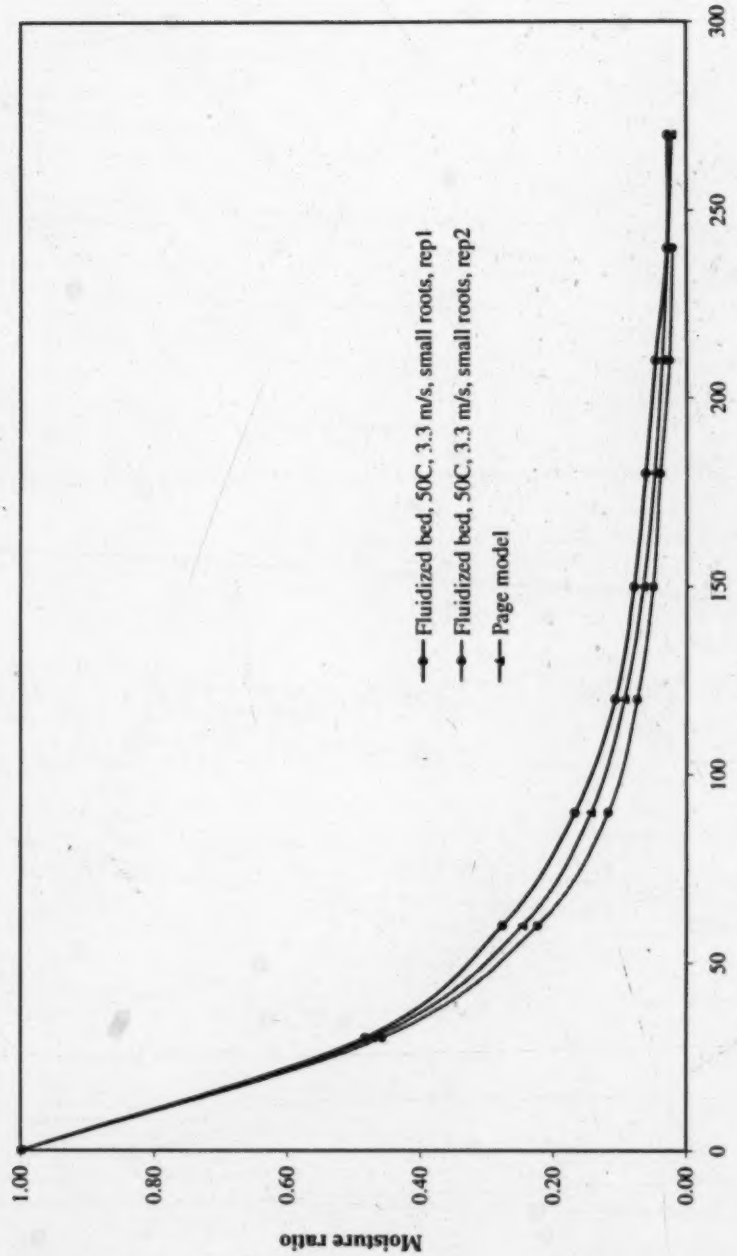


Figure 2. 19 Fitting Page model to fluidized-bed drying data

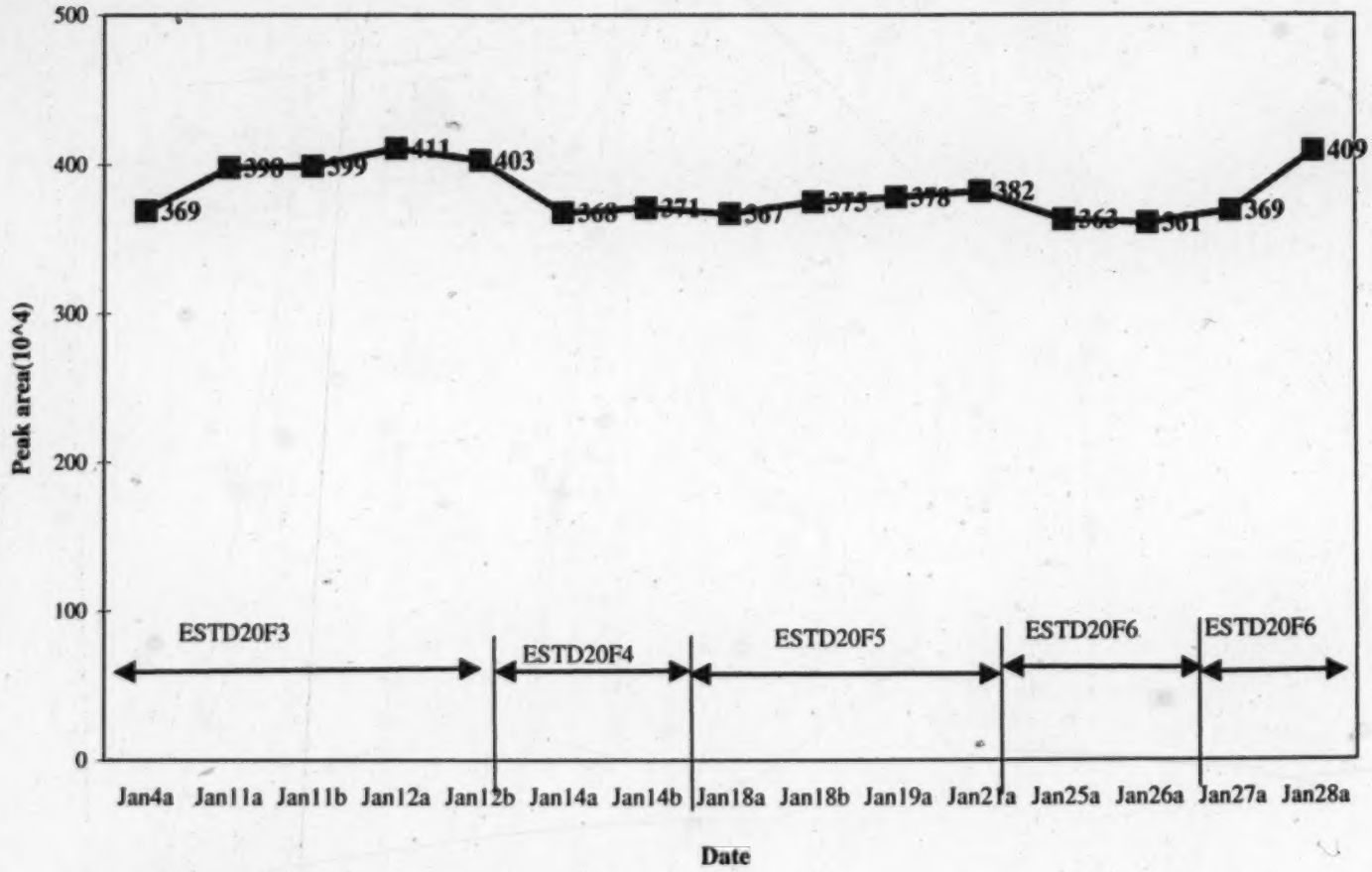
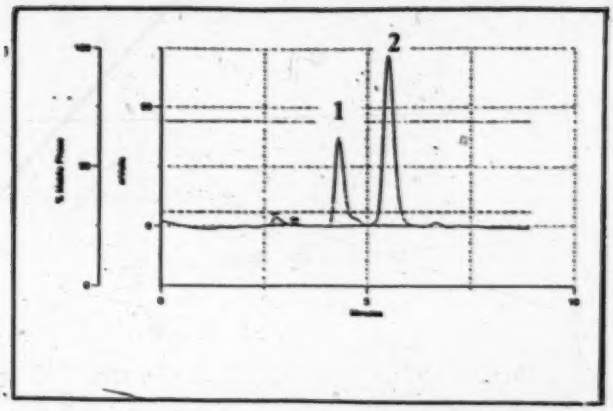
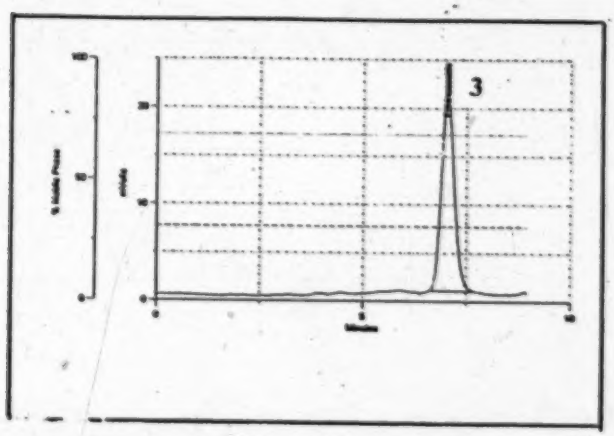


Figure 2.20. Echinacoside standard control chart

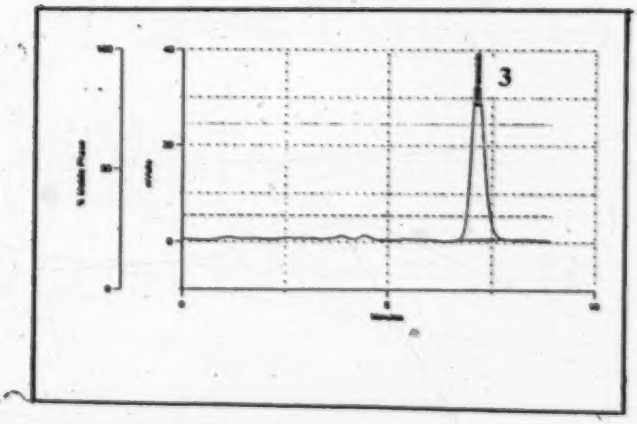
(a)



(b)



(c)



(a) Fraction 1 (b) Fraction 2 (c) Fractrion 3 3: Echinacoside

Figure 2.21. Typical chromatograms of echinacoside HPLC analysis

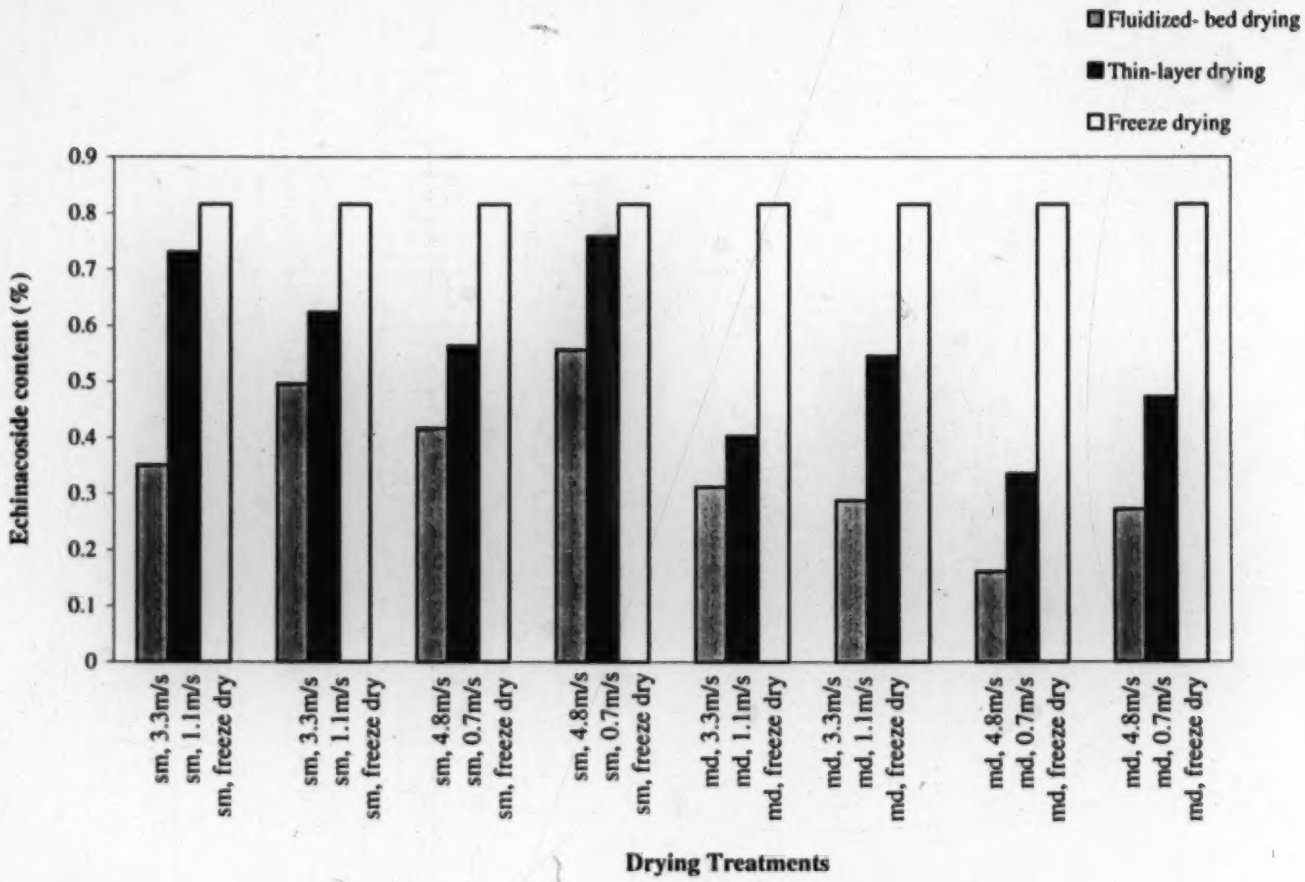


Figure 2.22. Comparison of echinacoside retention after 30C drying (sm=small, md=medium)

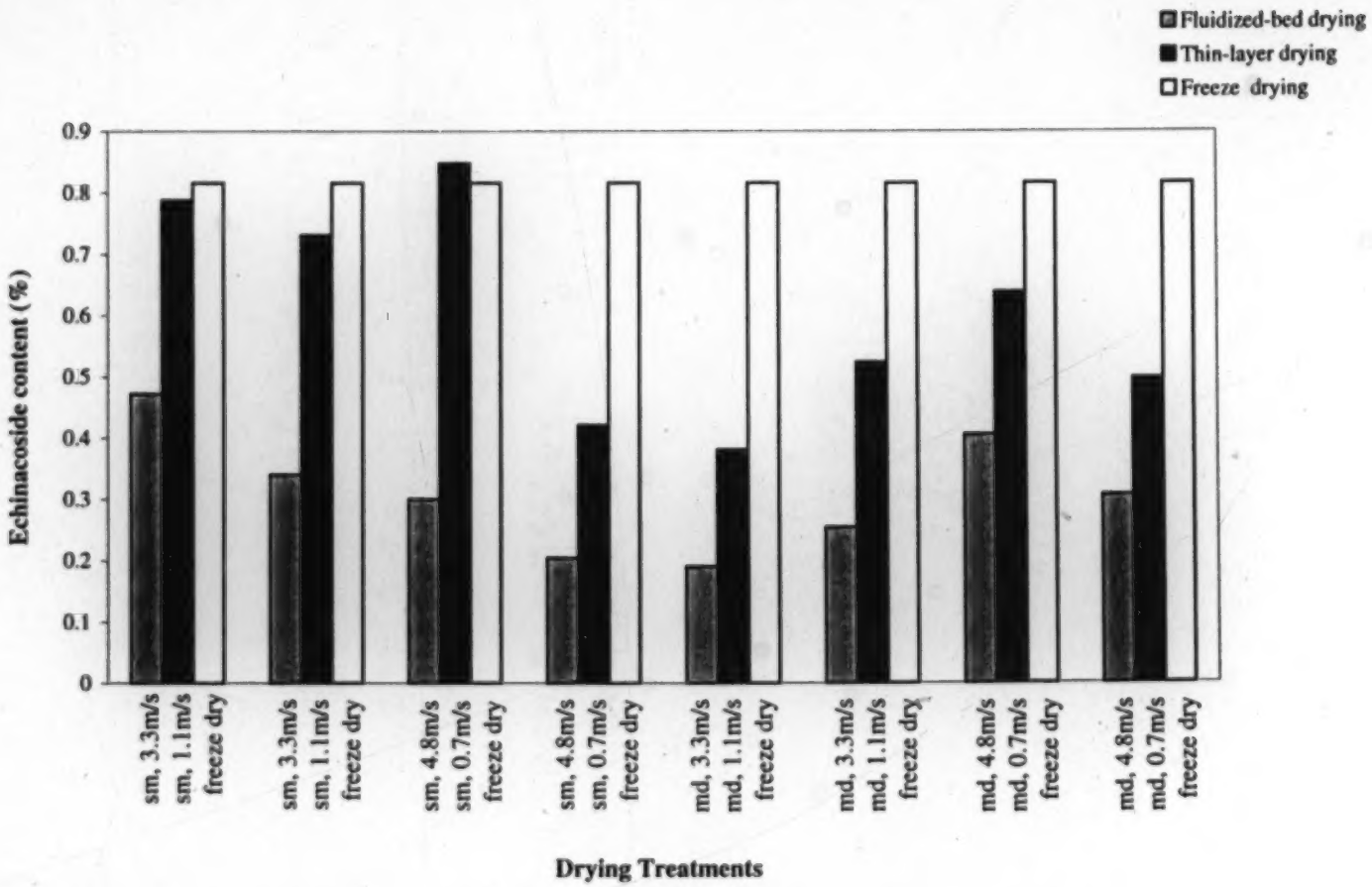


Figure 2.23. Comparison of echinacoside retention after 50C drying (sm=small, md=medium)

3. STORAGE STUDY ON DEHYDRATED *E. ANGUSTIFOLIA* ROOTS

3.1. INTRODUCTION

In contrast to the vast amount of literature information on nutritional value of fresh or processed foods, it is only in recent years that special attention has been directed to the effect of storage on food nutrients (Purvis, 1983; Sornsrivichai *et al.*, 1989; Bracy *et al.*, 1992; Sankat *et al.*, 1995; Naz *et al.*, 1997). The most important parameter affecting the nutritional quality of foods during storage is temperature. However, other factors such as packaging, atmosphere inside and outside the package, and exposure to light, may also be important for specific foods (Richardson, 1976). The medicinal values of processed medicinal herbs throughout shelf life is considered to be a very important element for herbal products (Bombardelli, 1991). Properly selected storage conditions will keep the active constituent degradation in herbal products to a minimum, and thus a prolonged shelf life with reliable quality can be achieved. On the contrary, a severe loss of active ingredients will occur if improper storage conditions are applied. Therefore, a long-term storage study is necessary before a medicinal herbal product is released to the market.

The storage conditions to ensure a high quality Echinacea product have not been clearly defined due to the complexity of quality standardization of this medicinal plant. However, it is certain that the processing procedure and the conditions of subsequent

storage will affect the active constituents of Echinacea products. As reported by Schulte *et al.* (1967), the polyacetylene content decreased markedly during long-term storage of fragmented Echinacea root material. According to Perry *et al.* (1999), levels of all alkylamides in *E. purpurea* roots decreased by over 80% during storage at 24°C for 64 weeks. They concluded that most herbal medicines containing *E. purpurea* roots would have much lower levels of biologically-active alkylamides than were present in freshly harvested materials. The same would be true for herbal medicines containing *E. purpurea* tops and *E. angustifolia* roots. Moreover, Perry *et al.* (1999) mentioned that the alkylamides from *E. purpurea* are stable in solution, and thus Echinacea tinctures may not suffer alkylamide instability problems.

Currently, the commercial Echinacea root powders in capsules and tablets are mostly freeze- or spray-dried (Hobbs, 1994). Those powdered roots might be more sensitive to their surrounding environment due to the larger exposed surface area and higher hygroscopic nature as compared to other types of Echinacea products. Bauer *et al.* (1988) have found that the polyacetylene compounds in powdered *E. pallida* roots can be readily oxidized. There is, however, little information on the stability of echinacoside in powdered Echinacea roots during long-term storage. The objective of the present study was to investigate the stability of echinacoside in powdered dry *E. angustifolia* roots stored under various conditions for six months. As a result, suitable packaging method and storage conditions, which can effectively preserve the echinacoside in powdered roots, were recommended.

3.2. EXPERIMENTAL DESIGN

The *E. angustifolia* root samples for long term storage study were prepared by freeze-drying. Two temperatures of 23°C and 4°C, and two packaging methods of air-packaging and vacuum-packaging were selected as test variables in this study. As a quality marker, echinacoside in root samples were monitored at day 0, the 1st week, and monthly thereafter up to six months. For each particular storage condition, two replicate storage samples were prepared. The echinacoside in each sample was analyzed in duplicate following the analytical methods as described in Section 2.4.1.

3.3. MATERIALS AND METHODS

Five kg of cleaned raw E. *angustifolia* roots were frozen at –20°C until used in the storage study. The frozen roots were then freeze-dried in a pilot freeze dryer (Virtis Co. Inc., Gardiner, NY) for 48 h. The freeze-dried roots were ground in a Waring commercial blender (Waring Products Division, New Hartford, CT), and the powdered root was then thoroughly mixed in a TURBULAR shaker (Type T2C Nr 771223, Willy A. Bachofen, Switzerland) until the sample was homogeneous. Ten grams of powdered roots were taken from this homogeneous sample pool and the echinacoside analysis was performed in triplicates on the same day as sample preparation. The mean value of the triplicates was recorded and reported as the initial echinacoside concentration of samples for storage study.

The samples for storage were prepared as follows. Approximately 20 g root powder was taken each time from the sample pool using a 25 mL measuring cup. The samples were then packed into 4 mil polyethylene zip sample bags (15×10 cm) or mylar/PVDC/polyethylene vinyl/alcohol high barrier film bags (20×10 cm, UniPac Inc., Edmonton, AB), individually. The transmission rates of oxygen and moisture vapor in the high barrier film are 8.7 g/m^2/24 h, and 7.7 cc/m^2/24 h, respectively. The high barrier film bags were vacuumed and sealed by Multivac packaging machine (AG500, Sepp Haggenmüller KG, Germany). The polyethylene bags were simply zipped, allowing the free transmission of ambient air into bags. The air components in air-packaged bags were measured randomly by a Basic™ Gas Chromatography (GC 8700, Carle Instruments, Inc., NY). The average percentages of O_2, CO_2, and N_2 of air in the bags were 20.6%, 0.068%, and 79.3%, respectively. The vacuum-packed and air-packed samples were equally divided into two groups. Each group of samples were placed separately in a walk-in cooler (4°C) and a box in the laboratory drawer (23°C), where light was completely eliminated. During the storage period, the samples were periodically inspected to ensure that all packages were properly sealed and desired temperature was maintained in storage environment. After the first week of storage, 8 bags (two bags under each storage condition combined by each type of packaging and temperature) were removed from the storage locations simultaneously. The echinacoside analysis as described in Section 2.4.1 was carried out in duplicate for the sample in each bag. The mean values of echinacoside, along with the storage conditions and storage time were recorded for further statistical analysis. Same sampling and

analytical procedures were followed every month until the 6-month storage experiment was completed.

3.4. DATA ANALYSIS

The effects of temperature, packaging, and storage time on echinacoside content, as well as the interactions between those factors were determined by performing a three-way analysis of variance using General Linear Model (GLM) procedure provided in SAS Statistical Software (SAS, 1998). Means of the individual factor were compared using the ESTIMATE function of SAS. The multiple comparison of interaction means was performed using ADJUST PDIFF function of SAS. The trend lines for echinacoside change under specific storage condition were generated in Microsoft Excel 97.

3.5. RESULTS AND DISCUSSION

A significant ($p<0.01$) change in echinacoside content of powdered *E. angustifolia* roots was found during the 6-month storage period. The maximum echinacoside loss, after six-month storage was 20%, and could be higher if the storage was extended. The interaction between the storage time and temperature was insignificant ($p>0.05$), while the interaction between storage time and packaging method was highly significant ($p<0.01$). This result suggests that the echinacoside in powdered Echinacea roots changes with time, but the trend of change varies with the two different packaging methods employed in this study. The changing trends of

echinacoside content in the vacuum-packaged and air-packaged samples are presented in Table 3.1. The echinacoside in powdered Echinacea root was well preserved during the first 3 months of storage. No significant ($p>0.05$) echinacoside loss was found during this period for both the vacuum- and air-packaged roots. However, the amount of echinacoside in air-packaged roots started to decline significantly ($p<0.05$) after the third month storage. This indicates that the third month may be a very critical point for the storage of powdered Echinacea roots. The powdered roots with vacuum packaging were stable during the first 5 months of storage without any significant ($p<0.05$) change of echinacoside content. However, a significant ($p<0.01$) decline of echinacoside was obtained in vacuum-packaged roots at the sixth month. A significant echinacoside decline was also observed in air-packaged roots at the sixth month of storage ($p<0.05$). These observations imply that significant echinacoside decline might continue in both air-packaged and vacuum packaged roots with the extended storage. The different responses of echinacoside observed in the two packages demonstrate that the degradation of echinacoside during storage was largely induced by air oxidation. However, other factors may also be involved in this degradation considering the fact that echinacoside also decreased in vacuum-packaged samples at the six month storage. The trend of echinacoside change under the two storage temperatures (4° and 23°C) is shown in Figure 3.1. No significant ($p>0.05$) effect of temperature on echinacoside change was observed during the 6-month storage period ($p>0.05$). As well, the interaction effect between storage temperature and time was not significant ($p>0.05$). Therefore, storage at room temperature might be acceptable for powdered Echinacea root.

3.6. CONCLUSION

Echinacoside in powdered Echinacea roots is relatively stable. It can be maintained at a satisfactory level for 3 months by air-packaging with 4 mil polyethylene bags. However, to prevent the echinacoside from further degradation over 3 months, vacuum packaging with a high barrier film would be necessary. A significant decline of echinacoside content can occur even in vacuum-packaged roots if the storage is extended beyond 5 months. Therefore, powdered Echinacea products stored over 5 months may have poor medicinal property due to the echinacoside loss during storage. Echinacoside in the powdered roots did not appear to be affected by storage temperature. Therefore, room temperature storage might be acceptable for the powdered Echinacea roots with respect to echinacoside preservation.

3.7. REFERENCES

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Table 3.1. Change in echinacoside content with two packages over 6-month storage (4° and 23°C)

Storage conditions	Echinacoside (% d.b.)							
	Day 0	1st Week	1st Month	2nd Month	3rd Month	4th Month	5th Month	6th Month
Air packaged	0.816[a]	0.814[a] (0.008)*	0.795[a] (0.009)	0.792[a] (0.013)	0.786[a] (0.015)	0.730[b] (0.025)	0.697[b] (0.011)	0.658[c] (0.018)
Vacuum packaged	0.816[a]	0.808[a] (0.008)	0.811[a] (0.005)	0.809[a] (0.008)	0.803[a] (0.010)	0.794[a] (0.006)	0.791[a] (0.018)	0.754[b] (0.013)

a-c Means within the same row with different superscripts differ significantly ($p<0.05$).
* Values in parenthesis are standard deviations, n=4.

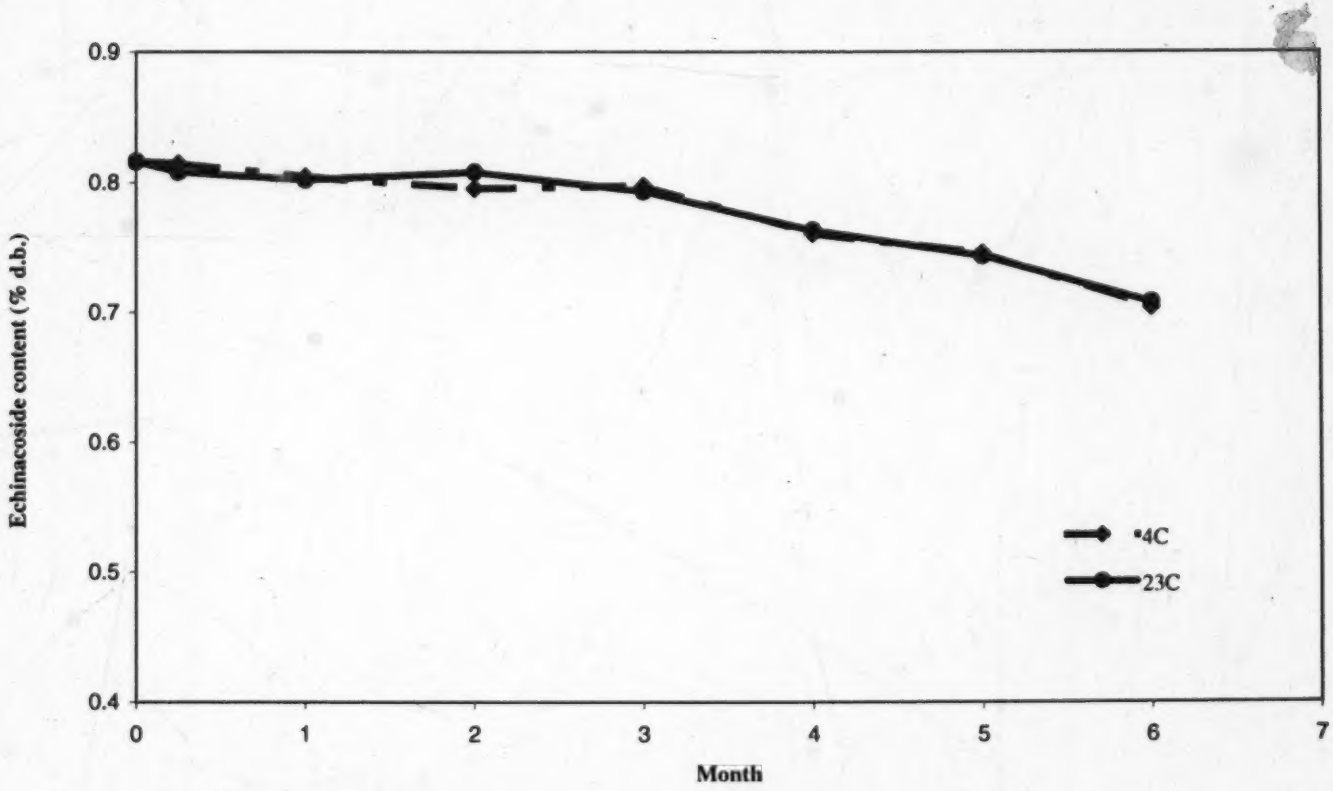

Figure 3.1. Change of echinacoside content at two storage temperatures

4. GENERAL CONCLUSIONS AND RECOMMENDATIONS

4.1. SUMMARY OF THE RESEARCH FINDINGS

Echinacea, a popular medicinal herb indigenous to North America, is one of the most promising immune strengtheners, with numerous scientific studies and rich clinical evidence in its favor (Cheminat *et al.*, 1988; Stotzem *et al.*, 1992; Jakic *et al.*, 1994; Erhard *et al.*, 1994; Burger *et al.*, 1997). There is, however, very little scientific information on Echinacea with respect to its processing, even though processing has been recognized as a crucial element for product commercialization. In this study, *E. angustifolia* roots were dehydrated using a forced-air thin-layer dryer and a fluidized-bed dryer under various drying parameters, such as air temperature, air velocity, and root size. The drying characteristics of Echinacea roots under different conditions were examined and the quality of the dried roots was evaluated by RP-HPLC analysis with echinacoside as a quality reference. A six-month storage study was conducted on powdered *E. angustifolia* roots and the quality of roots with respect to echinacoside content was evaluated. Based on the findings, suitable air-drying method, drying and storage conditions were recommended.

The work carried out in this research project reveals that for either fluidized-bed drying and thin-layer drying, the effects of air temperature and the size of roots on the drying rate were more pronounced than that of air velocity. The higher the air temperature or the smaller the root size, a higher drying rate was observed. Therefore,

increasing the air temperature within a certain range (30-50°C) might be the most effective method to increase drying rate when Echinacea roots are dehydrated in air dryers. Results of the study also suggest that the Echinacea roots with different sizes should be air-dried separately so as to avoid over-drying on smaller roots. It was demonstrated in this study that the drying of Echinacea roots occurred mainly in the falling drying rate period, where the mass transfer is primarily sub-surface controlled by moisture diffusion. This may explain the insignificant effect of air velocity on drying rate in both forced-air thin-layer and fluidized-bed dyers. Therefore, increasing air velocity in forced-air dryer is not an effective means to improve the drying rate of Echinacea roots. It was shown that fluidized-bed drying provided much higher drying rate than thin-layer drying under similar drying conditions. However, a considerably lower drying rate was observed in the fluidized-bed dryer shortly after the drying began. This might be due to the case-hardening of the roots as a result of a very fast moisture removal at the initial stage of fluidized-bed drying.

The dehydration procedure of *E. angustifolia* roots in both fluidized-bed dyer and thin-layer dryer can be described by the Page equation (Page, 1949). The drying constants (K, N) under various experimental conditions were calculated and correlated with air temperature, air velocity, and size of roots in both fluidized-bed and thin-layer drying. As a result, the duration of an air-drying process for Echinacea roots to reach a desired moisture level can be predicted as a function of air temperature, air velocity, and size of roots.

Under the experimental conditions of this research, thin-layer drying was found to be more effective ($p<0.05$) in preserving the echinacoside content than fluidized-bed drying. The severe loss of echinacoside in fluidized-bed drying might be related to the oxidation due to a large contact area between roots and air, the chemical modification due to the high rate of moisture loss at the initial drying stage, or the combination of these factors. The air temperatures and air velocities specified in this drying study were found to have insignificant effect on echinacoside retention. Therefore, a thin-layer drying, air temperature of 50°C, with 0.7 m/s air velocity appeared to be an effective drying process which can conserve energy and provide an acceptable echinacoside retention.

The results of storage study reveals that the quality of powdered Echinacea roots can be successfully preserved for 3 mo by air packaging with 4 mil polyethylene bags. To extend the storage over 3 mo, vacuum packaging is necessary. However, storage over 5 mo will result in significant ($p<0.05$) loss of echinacoside, even by vacuum packaging. No significant difference was found between storage at room and refrigerated temperature on echinacoside retention. Therefore, room temperature might be acceptable for Echinacea root storage.

4.2. RECOMMENDATIONS TO FURTHER STUDIES

From the aforementioned, a number of further research investigations may be recommended.

1. The potential of fluidized-bed drying for Echinacea roots should be further investigated by applying more suitable operational parameters. Case hardening of roots appeared to be a major drawback in fluidized-bed drying. However, case hardening can be corrected by modifying the physical properties of air, i.e. relative humidity (RH), and temperature. In this study, compressed air with extremely low RH was employed for fluidized-bed drying (particularly at 30°C). Most likely, the nature of compressed air applied in fluidized-bed dryer led to the case hardening development in Echinacea roots. A better designed fluidized-bed dryer with a moisture manipulating system might be necessary to further explore the potential of fluidized-bed dryer for this purpose. Also, a more suitable air temperature to provide sufficient drying and better echinacoside retention need to be determined for fluidized-bed dryer.

2. In this study, the *E angustifolia* roots were chopped into 1 cm length before they were air-dried. The small roots appeared to result in a higher drying rate and preservation of more echinacoside than medium roots. Therefore, it might be meaningful to investigate the drying behaviors and quality changes of Echinacea roots of different dimensions further. Elimination of case hardening maybe possible if the roots are cut to smaller size for fluidized-bed drying.

3. Quality of fresh roots stored for different periods of time under different storage regimes should be further investigated to determine the best way to preserve the quality of fresh roots before further processing.

4. Used as a quality marker in this research, echinacoside is actually one of the most important active ingredients in Echinacea species. Recent studies suggested that cichoric acid, some lipophilic components, such as alkylamides, polyacetylenes, and polar polysaccharide fractions are also very important markers for Echinacea quality (Wagner and Proksch, 1981; Bauer and Wagner, 1991; Bone, 1997). Therefore, future research attention should also be focused on the role of those constituents in Echinacea quality with respect to processing and storage.

5. Significant echinacoside declines was observed at the end of storage study in both vacuum- and air-packaged roots. An extended storage study might be necessary to further investigate the stability of echinacoside and evaluate suitable shelf conditions for Echinacea product.

6. Microbial quality of Echinacea roots, air-dried under different conditions, should be evaluated in future studies.

4.3. REFERENCES

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Appendix I. Drying constants and echinacoside loss in each drying run

Sample Code[1]	K	N	Apparent echinacoside loss[2] (% d.b.)
1T5011S	0.015285	0.946858	0.044 0.012
2T5011S	0.012292	1.032172	0.069 0.101
1T5007S	0.009140	1.042960	0.364 0.426
2T5007S	0.013667	0.947846	0.041 -0.105
1T5011M	0.008231	0.971530	0.46 0.41
2T5011M	0.010036	0.967931	0.286 0.299
1T5007M	0.009041	0.976801	0.224 0.133
2T5007M	0.008302	1.008187	0.321 0.318
1T3011S	0.010206	0.902892	0.186 - 0.016
2T3011S	0.011145	0.876452	0.197 0.187
1T3007S	0.009221	0.894680	0.069 0.045
2T3007S	0.009171	0.913686	0.336 0.169
1T3011M	0.009334	0.872480	0.256 0.285
2T3011M	0.008145	0.885382	0.336 0.496
1T3007M	0.004380	0.943025	0.505 0.455
2T3007M	0.007555	0.881370	0.421 0.265

Sample Code	K	N	Apparent echinacoside loss (% d.b.)
1FL5033S	0.055193	0.765498	0.362 0.327
2FL5033S	0.045278	0.846187	0.509 0.444
2FL5048S	0.059840	0.733413	0.445 0.587
3FL5048S	0.046216	0.803144	0.613 0.61
1FL5033M	0.042894	0.789824	0.621 0.632
2FL5033M	0.041860	0.789790	0.558 0.567
2FL5048M	0.042827	0.753723	0.512 0.509
3FL5048M	0.052548	0.733239	0.489 0.386
1FL3033S	0.073219	0.597102	0.335 0.306
2FL3033S	0.028648	0.778229	0.483 0.447
1FL3048S	0.069106	0.640276	0.092 0.026
2FL3048S	0.024963	0.819443	0.365 0.433
1FL3033M	0.027900	0.759949	0.487 0.522
2FL3033M	0.052132	0.643441	0.479 0.577
2FL3048M	0.043610	0.665679	0.611 0.7
3FL3048M	0.064371	0.617184	0.508 0.58

[1]Explanation of symbols (Appendix II)

[2] Apparent echinacoside loss (Echinacoside $_{\text{freeze dried roots}}$– Echinacoside $_{\text{air dried roots}}$)

Appendix II. Explanation of symbols in thesis

Symbols used in the drying experiments represent the drying method and conditions used.

Example 1:

1T5011S

1 = Drying replicate 1
T = Thin-layer drying
50 = 50°C drying temperature
11 = 1.1m/s drying air velocity
S = small roots

Example 2:

1FL3033M

1 = Drying replicate 1
FL = Fluidized-bed drying
30 = 30°C
3.3 = 3.3 m/s drying air velocity
M = medium roots